23 January 2021

Dear Editor:

I have undertaken a thorough minor revision of the manuscript, addressing all of the reviewers' points, following many of their suggestions. I have done more, by adding a substantial Section 3.4 to provide analysis and simulations to show how grain-size fluctuations could *create* prominent, *non-migrating* impurity signals. This section not only covers a topic that interests Reviewer 2, but also goes much further than that. The findings of my study of are thus enriched; its core arguments and conclusions are unchanged. Relevant parts of the Abstract, Introduction (Sect. 1) and Conclusions (Sect. 4) have been adjusted to coordinate with this addition.

A new Fig. 1 (examples of ice-core records) and Table 2 (mathematical symbols) have been added, following Reviewer 1's suggestions. The original Figs. 1 to 10 are renumbered 2 to 11.

Fig. 12, Fig. S3, Movies S6 and S7 have been added to accompany the new Section 3.4 reporting the findings about the potential impact of grain-size fluctuations.

Movie S8 has been added to illustrate how a signal in an ice core during cold-room storage would change in time. It is used by the last Conclusion paragraph in Sect. 4.

The Supplementary File and the Data Repository have been updated for the new figure and movies. See: https://figshare.com/s/8607e837455c5188c207

My detailed response and description of changes are written below. The reviewers' comments (from RC1, RC2, RC3) are shown in blue.

On page 3 of the revised manuscript, you will see a footnote, referred to on Line 67. I read the TC guidelines: "*Footnotes should be avoided in the text, as they tend to disrupt the flow of the text. If absolutely necessary, they should be numbered consecutively.*" In this instance, I believe that the footnote is an absolutely-needed case as its content would disrupt the main text if embedded in it (if not as a footnote). The information is peripheral but of interest to specific readers, and I think that keeping it as a footnote allows the main text to flow well.

Following TC instructions, a marked-up PDF file showing all changes (in MS-Word track change) is attached to the end of this response file.

Thank you for your attention.

Best wishes,
Felix Ng

<u>Detailed response and description</u>

L = line number in revised manuscript

RC1: Anonymous Referee #1

The paper by Ng tackles an issue from a theoretical point of view that bothered ice core science for nearly two decades, i.e., the potential signal migration of bulk chemical signals through the vein network in the presence of an in situ temperature gradient. This hypothesis was put forward by A. Rempel in 2001, but while no unambiguous ice core evidence for this hypothesis was provided in the meantime it could also not be refuted as the theory of migration through the vein network is physically not disputed. However it is still unknown how much of the impurities are located in veins and, thus, really subject to this transport.

The results by Ng provide a convincing argument why no evidence for a migrated signal CAN be found, i.e., because any migration signal would also be subject to intense diffusional smoothing. Ng expands on the theory by Rempel by taking into account the curvature effect on vein volume that had been intentionally neglected by Rempel. Ng is able to convincingly show that taking this additional term into account, does not stop any migration but leads to strong diffusion of such a migrating signal and, thus,

to the disappearance of the displaced signal in the ice. This has two very important implications for ice core science: Firstly, any short-lived peaks in the ice core record that are imprinted in the veins cannot survive the migration process. Vice versa, if there is a peak found in the ice core record it cannot be shifted relative to its initial bulk position. Secondly, the presence of distinct peaks in many deep ice core records suggests that vein migration and diffusion cannot be a dominant process as otherwise any peaks would have disappeared. This implies, as explicitly outlined in the paper by Ng, that either the part of impurities located in veins is only small or that vein transport is increasingly suppressed by a fragmenting vein network as already suggested by other authors.

The paper is well written and excels by its stringent, mathematical approach, while at the same time performing model experiments that are very instructive and, therefore, of great value for the ice core practitioners. It is therefore of high relevance and well suited for publication in The Cryosphere. The mathematical approach (although well laid out) and some for the outsider unintuitive formulations make the access to the paper a little more difficult than necessary. Below I make some suggestions for minor revisions that may help to remedy that. In summary, I highly recommend the paper for publication in The Cryosphere after taking care of these minor revisions.

I thank the reviewer for the appraisals and the valuable suggestions below.

General comments

The paper is a little "dry" as there are no ice core examples provided. Thus, for those readers who are not specialists in ice core chemistry, it is hard to imagine how those signal that Ng talks about, look like. Here, it would be helpful to see (in the Introduction or the Discussion) a figure that shows examples for high peaks in deep ice on the one hand (for example for in the Ca2+ or SO42- record) and examples of a general reduction of variability as seen on others (for example in the Na+ and NO3- record). Examples can be found in the papers by (Traversi et al., ES&T 2009, Röthlisberger et al., CP 2008, Schüpbach et al., Nature Communications 2018, Barnes et al., JGR 2003, and others).

The new Figure 1 has been added to show ice core examples, from Antarctica and Greenland. Thank you – this addition really enriches and embellishes the paper.

As mentioned in the interactive discussions, I do not evaluate the variability/trends observed on such records, and leave that to the observational / future studies. The focus is to illustrate the abundance of ionic peaks and their expressions and variety in deep ice. Thanks for pointing me to Schüpbach et al. (panel c). Also, R. Traversi kindly provided data to me, allowing me to make panels a and b. And I managed to find a published piece of high-resolution data for NGRIP (panel d). Other than this, I have not found usable data, as publicly available/archived chemical data at a resolution of 50 cm or better are scarce. A resolution as high as 10 cm is necessary to portray the peak forms for my purpose.

L53 and L331 now refer to Figure 1.

In addition to Table 1, which provides the values of the constants used, I would suggest to provide a table 2 where all variables used are listed with a short explanation and their units.
Done. Table 2 has been added. To avoid a very long table, I restrict it to the model variables appearing in the main text, and exclude those variables local to Appendices A and B.

The information on the age span is provided in figures 6 and 7, however the discussion of this parameter, despite its great importance for ice core science in particular for very old ice, is rather limited. Looking at the EPICA Dome C results it appears that the age span in Fig. 7 approaches several thousand years or even the precession age scale when reaching an age of 700-800 kyr (unfortunately the current scale of the x-axis does not allow to quantify this for the control run). On the other hand, the EPICA Dome C ice core record provides some information about the time scale of variability that can still be resolved, thus, would constrain the degree of vein transport smoothing empirically. I would recommend to extend the discussion on this point, as it is of great relevance for ice core sciences.
<u>Age span (revised paragraph on L366 to L380):</u>
Discussion has been added on L370–373. I appreciate your interest in seeing the computed age spans being used to interpret the actual details of the EPICA ice core at depth. However, I caution against doing so, giving a specific reason on L372-373, and adding another overarching reason on L377–380. The opening sentence of the paragraph (L366) now includes “age span” as a signpost to the paragraph’s topic. Note that the old Figs. 6 and 7 are now Figs. 7 and 8.

The horizontal “age span” axes in Fig. 8c and 8f are not scaled for the control runs (their axes focus on the other runs) because in deep ice, the signal amplitudes have diminished to near zero, as shown in Figs. 8a & 8d. I have added a final sentence in the caption of Fig. 8 to explain this (L1005–6).

Specific comments:

Introduction 1st paragraph. You could also mention here already the loss of species such as NO3- or Cl- at the surface (Röthlisberger et al., Ann Glac 2002, Weller et al., JGR 2004) migration processes (for example for methanesulphonate) that occur already at the surface (Osman et al., 2017) or the aggregation of dust particles in the deepest ice (Tison et al., 2015)
I have not done this. I prefer a more focussed thread leading into the core matter of the study (2nd paragraph onward) and feel that adding these details to the 1st paragraph would detract from that.

p2 l33: "...migrate relative to the ice..."
Done; L33.

p2 l35: "... could decouple..."
Done; L36.

p2 l41: "... signal migration in deep ice may..."
On L42, I have not followed your suggestion to write “in deep ice”, because signal migration (the process) can occur --- and be limited --- at any depth. (It is true that large displacements manifest at depth as a result of signal migration.) I therefore prefer the existing wording.

p2 l51: this is one of the examples where the author refers to the ice core evidence without showing it
Thank you. The new Figure 1 showing ice-core records is now referred to on L53 in a new sentence. I adjusted the wording on L52 to coordinate with this.

p2 l62: One intrinsic assumption made in the Rempel theory and also in the work by Ng is that c_B=c*Phi. However, if impurities are located in microinclusions or grain boundaries, hence not in contact with the veins, then the vein transport is not representative of changes in bulk concentrations. In fact, this is discussed later, but I would recommend that this assumption is explicitly mentioned when c_B=c*Phi is introduced.

Yes, I now mention this assumption explicitly by:
(i) clarifying the definition of c_B (including its units) on L58–60, and
(ii) writing “With c_B encapsulating vein impurities, the relation c_B = c*phi holds…” on L66–67.
Alongside these changes, an opening phrase on L105–106 has been modified.

In the caption of the New Figure 1, I also clarify that the impurity concentration of an ice-core record is the total concentration, whereas c_B refers only to the vein impurity component (L868–869).

p3 l63: when I first read the word "ice porosity" I got confused. I would suggest to write: " is mirrored by variations in the liquid filled vein volume relative to the total ice volume, in the following called ice porosity Phi"
On L66, I now write “porosity” instead of “ice porosity”, and I define this term in the same sentence by writing

> “… in the porosity (Fig. 2c, d), which represents the volume fraction of veins in the ice.”

p3 line 68: please change the unclear wording "Thus the c_B peak translates"
Done. Changed to “Thus the peak signal in c_B translates”; L71. (On L71–72, in order to be informative, I added a phrase in brackets to say that “the same translation applies to trough signals”.)

p4 l96: replace "we" by "I" throughout the manuscript
I prefer not to do this in this manuscript. I understand that some scholars prefer what you suggest.

p4 l111: "The three terms on its right-hand side describe the temperature depressions due to (i) solute, (ii) interfacial curvature (the Gibbs-Thomson effect) and (iii) pressure, respectively;"
Done; L114–115.

p4 l 113: what does the constant gamma represent?
Now clarified on L116 (gamma is interfacial energy). I adjusted the entry for gamma in Table 1 to coordinate with this change.

p6 l147: "... where the melt rate m (in units?) at the interfacial boundaries of the vein accounts..."
Done. On L153–154, I have clarified these in two linked sentences.

p6 line 155: why not explain the last term here?
Revised. In the old paragraph, the reader was meant to understand that the last term describes vein motion (which was treated previously, for porosity), but the paragraph’s structure did not pull this off well enough. I have now rewritten the first 3 lines of the paragraph so that which term corresponds to which process is clear. Please see L160–162. I also remind the reader that the vein motion is the same vein motion that was considered a few paragraphs ago (L161, phrase in brackets).

p7 equations 17 and 18: You neglect dw/dz. Say explicitly why.
Done; L178. I say here that incompressibility (Div.u = 0) has been used in deriving this result (dw/dz wasn’t neglected; dw/dz + du/dx + dv/dy sums to zero). This clarification on L178 is paired with an earlier clarification on L170.

p11 l256: "while anomalous (Rempel) diffusion"
Done; L265.

p11 l273-274: use a different variable for the basal melt rate (for example m_bas) to distinguish it from the interfacial melt rate m used above
Done; L283–284.

p12 l298: Likely a topic to be looked at in a separate paper, but the theory by Ng should be also able to predict how much vein diffusional smoothing occurs for the water isotopes.
Thanks for this suggestion. It is best left to a separate paper, and I haven’t modified the text, because while one can consider diffusion of (i) signals of water-vein ionic species and (ii) signals of isotope abundance, some key physics differs between (i) and (ii). Topic (i) involves the solute-controlled liquidus, while (ii) doesn’t. Topic (ii) involves equilibrium fractionation but (i) doesn’t. There are broad parallels but I expect the model equations to be rather different.

p13 l318: " to become so large to be ..."
Thanks; done; L328.

p13 l321. The data provided in the Mayewski papers are not of really high resolution.
You may also want to cite Schüpbach et al, Nature Communications 2018, Bigler et al., Quaternary Science Reviews 2010, Röthlisberger et al, CP 2008)
Thanks for pointing this out. On L331–332, the Mayewski references have been removed. I have added your suggested references (except Bigler et al., whose study extends to 2 km depth only and thus somewhat marginally illustrates my sentence) and included a reference to Svensson et al. (2013), whose data feature in Fig. 1d. Also, on L331, I refer to the New Figure 1.

p13 l332: "we observe an interesting"
Done; L342.

p13 l335-337: expand the discussion of the age span and the potential resolution loss
Done. Please see my earlier detailed response to your last *General Point* above. The revised paragraph is on L366 to L380.

p15 3rd paragraph: The movies provided are very helpful!
Great to know!
In Movie S3 and Figure 9 one of the two peaks in the GRIP ice core moves relatively upward. This deserves some discussion in the main text.
Done; L388–389. Here I clarify that the movement is due to vertical ice compression.
Also the y-axis for the temperature profile in the movies could be scaled equally for all time steps.
I haven't done this for the movies (e.g. Movie S3) because the temperature curve would still jump from frame to frame, as MATLAB (my plot-making software) puts its 'tickmarks' to span the y-axis so that the top and bottom tickmarks always lie at the corners of the plot. I am not enough of a "Matlab guru" to know how to control the plot element properties to be able to overcome this.

p16 l403-404. I am not sure I understand this correctly, please clarify. Again, a discussion of the time-scale that could be resolved and those found in ice core records would be helpful.
Done. Please see L419–421. For the clarification part, I have rewritten the Original L403–404 as

"it is understood that fewer high-frequency palaeoclimatic details are retrievable from deeper ice, due to the finite resolution of ice-core sampling, alongside layer thinning, which causes more time to be encapsulated in a given ice thickness."

I have not discussed the time scale that could be resolved, because of my reservations (expressed before) about using the model-computed age spans to analyse the details of real ice-cored records, in deep ice (see my earlier responses on "age spans"). The analysis of real records falls outside the scope of the present study and I feel that this is best left to another piece of work.

p17 scenario 2: here you mention the issue of micro-inclusions which would contradict the assumption that c_B=c*Phi. As mentioned before, this assumption should be qualified as such earlier in the manuscript.
Yes. In response to your earlier point for "p2 l62", the assumption behind c_B = c*Phi and an explicit definition of c_B have now been given early in the manuscript. Also, I have now made several minor wording changes in Section 4 so that whenever the symbol c_B is used, it refers only to the vein impurity component, whereas the *measured* impurity records quantify the total concentration --- including vein, grain-boundary and matrix contributions. The corresponding changes can be found on L495-6 (bracket), L511, and L532.

p17 l455: "vein c_B" is the wording correct???
Reworded now as "the signals in c_B"; see L543.

p17 l458-459. The sentence "In any case... from SO42-" is way too general and should be specified. Fr example to which depth could we see volcanic peaks at GRIP and EPICA Dome C. Which longer-term variations could be still resolved (discussion of age span)
Yes, on L548–550, I have now removed the reference "… *as done in volcanic flux reconstructions from SO42-*" and rewritten the sentence – in order to be specific – as:

“These considerations caution against interpreting all observed ionic signals directly for palaeoclimatic events and variations: some signals may be distorted in form and duration, and some peaks may be caused by local grain fining (this may result from recrystallisation processes (Faria et al., 2013) or high levels of dust/microparticles in the ice (Alley et al., 1986a)).”

As explained above, it is beyond this paper’s scope to address the specific details of measured records with the computed age spans.

p18 l2: "of any vein impurities"
Done. Replaced “all” by “any”; L554.

p18 line 476-478. expand the discussion on longer variations
I have deleted this short paragraph from the middle of Section 4. The longer variations had already been discussed in the final paragraph of Sect. 3.3 (L414–423) and I have no more to add here.

p19 l498-500: This comes unexpected and justifies a little bit more discussion.
Yes. The paragraph at the end of Sect. 4 was a little abrupt: its brevity didn’t help. To inform readers more, I have extended it by describing how fast a signal in c_B would diffuse/decay at two storage temperatures (see L591-596; L593–596 are new). Movie S8 is added to show the simulated results.

Figure 1: I know these are only illustrative figures, but the scale of the anomaly (centimeters) and the approximate location within the ice sheet of the sketch (lower third, where temperature gradients exist both in Greenland and Antarctica) should be indicated either in the figure itself or at least in the caption.
Done (note that Figure 1 has been renumbered as Figure 2). I followed your second suggested option. To the caption of Fig. 2, I added a sentence to describe the whereabouts of these gradients in an ice sheet, referring also to the actual examples shown in Fig. 4; see L889–890. Now L890-891 describes the scale of the signal/anomaly in panels b, c and d, without pinpointing where though, because such signals can occur anywhere in the ice column.

Figure 2: clarify what you mean by "that measures distance from ice at age t"
Done. I now clarify this idea in two places:
- caption of Fig. 3 (Old Fig. 2), L902–905 have been expanded to clarify the idea;
- L252–253; rephrasing done here to elaborate the idea.

Figure 7: rescale panel f to show the age span for ice with an age of 800 kyr.
[Note: this figure is now Figure 8.] As explained earlier in my response, I do not rescale the age-span axes here, because the signals in the control run have decayed to near-zero amplitude at depth, but on L1005–1006 I have added a final caption sentence to explain the choice of axis scaling.

Reviewer #2 (RC2 and RC3)

Again, I thank this reviewer for his appraisals and suggestions.

Rather than to recount my interactions with Reviewer 2 during the *Interactive Discussions* process, here I summarise all of what I have done to manuscript following those interactions.

Those interactions, while valuable, have been mostly confined to one idea raised by the reviewer (and the surrounding considerations) --- his idea of how the Rempel theory might be justified under specific conditions. I think that this topic is useful, but has a limited value for the manuscript and for readers.

I went much further than covering that topic. In a New Section 3.4, on page 16–18, I analyse the model more and report what happens to cB if we <u>*prescribe*</u> a grain-size fluctuation in the ice. That is, fluctuation in d_g. This includes a short piece of mathematics (L455–466) to explain the interactions, and a set of simulations showing the results (L467–L488). The fact that these preliminary/artificial experiments do not involve a physically-based model of grain-size evolution (because this is out of reach) is again made clear (L425–430, L471–472).

The discovery is that a grain-size fluctuation can cause a new signal in cB to form, and that *the resulting signal is locked to the fluctuation and does not migrate relative to the ice*. Section 3.4 is accompanied by the addition of Fig. 12, Fig. S3 and Movies S6 and S7. These results extend the paper's findings. To coordinate with them, I have adjusted various wording and inserted qualification/signposts elsewhere in the manuscript:

- Abstract (L15–16, 18–20, 21–22 & 23, reporting and qualifying findings in regard to d_g fluctuations)
- Introduction (L55, signposting the new result)
- Model section (L135–7, L215–6, L248–9, sentences added to signpost later work with d_g)
- Discussions (L495–500, qualification added "*unrelated to grain-size fluctuations*")

(L514, 520–521, 527–531, 536, 546–547, 549–550, 552–553, 560, 569–570, 583, 589, embedding of the new results concerning d_g and their ramifications).

Early within the New Section 3.4, I also carried out my plan given in AC2 and AC3. On L430–L440, I added a paragraph to describe the concept --- raised by Reviewer 2 in RC2 and RC3 --- that a specific inverse-square coupling exists between mean grain size d_g and vein impurity concentration c_B to keep vein radii r_v uniform, so that the Gibbs-Thomson term is kept uniform and this suppress the Gibbs-Thomson diffusion. In the next paragraph, on L441–L454, I evaluate this idea, noting my reservations of it by considering its mathematical, empirical and theoretical bases. I think that these two paragraphs of 500 words suffice to address this topic which interests Reviewer 2.

Regarding Reviewer 2's idea that the observed anticorrelation between *total* impurity loading and grain size (d_g) in ice cores justifies the necessary coupling between c_B and d_g, I remain unsure. This is because the *total* measured loading (for an ion species) generally consists of 3 contributions: (i) impurities in veins, c_B, (ii) impurities at grain-boundaries, and (iii) impurities in the ice matrix/crystals, e.g. in microinclusions. Suppose we have measurements of a total quantity T = X + Y + Z, but have no measurements of X, Y, Z. The observation that "*T is inversely correlated with another variable P*" does not mean that "*X is inversely correlated with P*". Much as I recognise such statement as a possibility, it is not supported. With the currently available evidence, this is as far as I can go in terms of evaluating this particular idea --- please see L445–448.

Below, I copy and paste all of the text of RC2 and RC3, but have not written point-by-point response against it, as my overall response is given here.

RC2:

The manuscript by Felix Ng revisits the question of what happens to soluble impurities in glacial ice over the long time periods represented by ice-core climate records. Assuming that these impurities are primarily contained within a connected liquid-filled vein-node network along three- and four-grain contacts (e.g. Nye and Frank, 1973), an earlier model predicted that the control of impurity loading on liquid content should produce spatial variations in the effective compositional diffusivity that would help to explain the long-term preservation of short wavelength (e.g. representing seasonal to decadal time periods) bulk compositional signals, while also leading to their translation relative to the surrounding ice (Rempel et al., 2001). This so-called "anomalous diffusion" phenomenon has proved very difficult to test, since the predicted rate of signal migration is very low (largely controlled by the shallow temperature gradients in polar ice) and firm constraints are rarely, if ever, available on the relative timing of compositional and ice-borne (e.g. oxygen isotopes) proxy deposition in the distant past. If the compositional signals themselves aren't altered in form, but only displaced a small distance relative to the ice with which they were originally deposited, what evidence is there to unequivocally demonstrate such subtle effects?

Ng's analysis predicts qualitatively different behavior in a slightly modified system, by focussing on the role of vein surface energy embodied in the Gibbs-Thomson effect, while assuming that the ice is characterized by locally uniform (but slowly growing) grain sizes. The models of anomalous diffusion that had been presented earlier had differed by employing a different assumption concerning the relationship between ice grain size and impurity loading. Measurements show that average grain size and impurity content are negatively correlated in ice cores (e.g., Gow and Williamson, 1976; Gow et al., 1997; Lipenkov et al., 1989; Azuma et al., 1999, 2000; Thorsteinsson et al., 1995; Durand et al., 2009). Rempel et al. (2001) highlighted this anticorrelation as consistent with the assumption that "the surface energy of curved interfaces acts to make vein radii uniform (Nye, 1989; Mader, 1992)", reasoning that "variations in [bulk impurity loading] must correlate with changes in the total length of veins per unit sample volume". Effectively, the idea was that by responding to impurity loading, grain sizes could adjust and prevent differences in vein radii from occuring.

By contrast, Ng treats the grain size and impurity loading as unrelated, implying that vein radii are initially in disequilibium when changes in deposition produce changes in impurity loading, so that the vein radii subsequently evolve towards uniformity. This equilibration of vein radii drives additional computational transport that adjusts the impurity loading, thereby modifying the form of the compositional signals and preventing their displacement relative to the ice.

The analysis by Ng is elegant and clearly presented, and represents a welcome addition to the literature that describes post depositional processes with potential to be relevant to ice core records. To better convey the departure from the earlier efforts to address this particular problem, it would be helpful to clarify the discussion surrounding the different assumptions regarding ice grain size. At present, the reason given by Ng for neglect of the Gibbs-Thomson effect in the Rempel et al. (2001) model is the scaling in the liquidus relation (line 122). However, the original reasoning provided by Rempel et al. (2001, p. 370) emerges from the assumption that an anti-correlation between grain size and impurity loading makes vein radii uniform, as predicted if grain size scales inversely with the square of bulk impurity loading in equation (7) of the current work, thereby rendering the second term in equation (8) spatially uniform as well.

Unfortunately, as noted by Ng around line 239, "reliable grain-size modelling remains out of reach", though impressive improvements have been made in recent years (e.g. Faria et al., 2014; Ng and Jacka, 2014). Nevertheless, no convincing treatment has yet been provided that is quantitatively successful at representing the causal mechanisms that produce the observed anti-correlation between grain size and impurity loading. The current work highlights yet another reason, in addition to more commonly evoked considerations of the effects of grain growth on rheology and fabric development, for the importance of addressing this challenge.

Under the impurity-independent grain size assumption, Ng demonstrates convincingly that short-wavelength compositional variations (with impurities confined entirely to the vein-node network) should not persist over multi-millennial time scales under typical ice core conditions. Clearly, the apparent preservation of such signals deep in the ice requires further explanation, and Ng provides considerable insight into various potential mechanisms that might resolve this conundrum. Ultimately, two scenarios are championed, with the first involving some mechanism for blocking vein transport, for example with dust particles (e.g. Raymond and Harrison, 1975), and the second relying on impurities being largely confined to the ice matrix or two-grain boundaries (e.g. Eichler et al., 2017). The model of Rempel et al. (2001) shows that a strong coupling between bulk impurity content and grain growth could provide a third mechanism by reducing or eliminating spatial variations in vein radius and greatly damping the influence of the Gibbs-Thomson effect on impurity transport.

Likely some elements of all three
scenarios contribute, and efforts to quantify post-depositional changes would benefit from further efforts aimed at unravelling their relative importance. Ng's manuscript represents a valuable, detailed exploration of the end member case in which grain size is independent of bulk impurity content. This clear and cogent analysis has important implications, and it is to be hoped that it will motivate efforts that provide even further constraints on how exactly post depositional compositional migration occurs in nature.

I note that Reviewer 1 has already provided detailed suggestions for minor wording changes and I have nothing significant to add.

RC3:

I agree that it would be helpful to signpost a later discussion on the importance of grain-size variations for determining how vein constituents behave. I would also welcome having the ideas surrounding the specific assumption regarding uniformity of vein radii in the Rempel et al. (2001) model attributed to a personal communication. However, I do not think that such a reference is necessary. While it is true that the matter was not discussed at length in that work, the reasoning was explicitly provided by the statement that "as the surface energy of curved interfaces acts to make vein radii uniform, variations in cB must correlate with changes in the total length of veins per unit sample volume", with the following sentence going on to note the qualitative support provided by observed anti-correlations between bulk impurity content (cB) and grain size.

The twenty years since we completed that work have seen tremendous advances in the community's ability to characterize the physical and chemical characteristics of ice cores at increasingly fine scales. Despite these advances, it is noteworthy that we still lack a quantitative mechanistic understanding for precisely how the anti-correlation that is commonly observed between cB and grain size develops. Your new model brings welcome attention to the consequences of that important issue. In your comment you: 1. highlight the importance of the detailed form of the anticorrelation for determining the fate of vein constituents, 2. champion the generality of your formulation in being readily adaptable to examine the affects of different grain size evolution laws, 3. assert the need for a mechanistic understanding of the role of impurity loading on grain size, and 4. emphasize the inability of existing theories of grain growth to address this problem. I think we have broad agreement on each of these points, which offer a clear motivation for filling these knowledge gaps and bolstering confidence in the integrity and resolution of these important paleoclimate records.

Beyond the observed fine-scale grain-size variations themselves, an argument in favor of the uniform vein radius assumption employed in the Rempel et al. (2001) treatment is the long-term preservation and apparent fidelity of fine-scale cB signals recovered from ancient ice. If, as in your model treatment, vein radius evolves to force diffusive impurity redistribution, then your analysis implies that either those deep signals are distorted from their original form, thereby compromising detailed paleoclimate interpretations, or instead their preservation might be attributed to one or both of the mechanisms that you suggest, namely: residence outside of the vein network under much warmer conditions than the eutectic temperatures of their solutions, or blockages that manage somehow to severely restrict vein diffusivity. The observed anti-correlation between impurity content and grain size would in any case remain unexplained. However, should this problem be addressed, the precise manner in which grain sizes respond to impurity content or perhaps some coincident variable (e.g. impurities on two-grain boundaries) could be accounted for in a refined treatment that extends beyond the locally uniform grain size case that is the focus of the example calculations in your paper. While we're each free to argue over the set of assumptions we feel to be most reasonable, whichever situation actually dominates is not currently known.