Response by the author detailing the revision of manuscript "egusphere-2024-1012"

15 August 2024

Dear Editor of TC,

Thank you for considering the revised manuscript and for posing a further idea for me to consider. Please find below my point-by-point reply to your comments. I took the opportunity to go through the manuscript once again, to spot any glitches. I made a few small changes to correct typos and refine local wording, as listed below, and have uploaded the corresponding revised manuscript to the system. I uploaded also the Supplement file again, even though it has not changed.

Best wishes, Felix Ng

ED: Editor comments

Dear Author,

Thank you for your thorough revision of your manuscript, and your careful and adequate responses to the Reviewers comments. Your revised version is almost ready to be accepted but I would like you to consider addressing the following point, if possible. Line numbers below are those of the tracked changes version.

Line 112, you mention "excess diffusion apparently occurs in those sections and not others". Lines 169-170, you briefly allude that impurity content would enhance grain boundary and triple junction thicknesses, favoring excess diffusion. Interesting data on this aspect is given in Thomson et al. (2013), whom you cite and note that they studied ice "under different conditions". This is a rather vague statement. Isn't the important point "under increasing impurities concentrations", as detailed in Thomson et al.? Further, in your discussion, lines 935-936, you come back to the possible relationship between excess diffusion, and increased impurities contents that would thicken veins and turn on excess diffusion.

Could not all this be substantiated with data? Chemical analyses for most cores you discuss are available. Would it be possible to investigate here a possible correlation between excess diffusion and impurity content? Even a partial investigation on a single core would be valuable.

Thank you for this observation. I too feel that such correlative study – comparing the pattern of excess diffusion and the impurity records along an ice core – is worthwhile and should be explored more.

A major unknown in such study is how the *bulk* concentration of each impurity (e.g. as measured by CFA analysis) is partitioned in the material between ice crystals, the vein network, and grain boundaries, and how this partitioning varies with depth down-core. The potential influence of impurity concentration on isotopic diffusivity described in those passages (Lines 170 and 200-201 (Thomson et al, 2013) and Lines 813-814, *"impurities migrate to grain boundaries and then to the veins, thickening them..."*, and thereabout) specifically refers to the impurities at grain boundaries, not to the impurity bulk total. Indeed, the partitioning is a limitation of the experiments of Thomson et al. (2013); see the caveat mentioned on Line 200: *"impurity concentration at grain boundaries was estimated from the bulk concentration as it cannot be measured directly"*.

Therefore, when depth records of excess diffusion and (bulk) impurity concentration are compared, one doesn't know the fraction of impurity concentration at grain boundaries; guessing it is unsound. This greatly hampers interpretation. Although LA-ICP-MS mapping can tell us the location of impurities, very few maps are currently available, only for small surface sections spanning a few cm's. Also, how such maps could be used to calculate the impurity fraction at grain boundaries from the total remains to be researched.

More generally, a correlative study needs to involve other factors besides impurities – temperature, grain size, vein-water flow velocity, and the down-core pattern of blockage/disconnection of veins. The last two factors are again major unknowns, as mentioned in the manuscript.

Jones et al. (2017) attempted a correlative study of the kind you suggested, for the WAIS Divide ice core, to query the origin of the anomalous excess diffusion at \approx 15–18 ka BP. They used the profile of diffusion length estimated from the power spectral density of the isotopic record, and involved the bulk records of several ions (calcium, sulphur, magnesium) and acidity; see their Section 3.3. They could not find any convincing correlation between the profiles that would explain the diffusion anomaly. One reason, described above and briefly alluded in their study, is that we do not know what part of the impurity concentration pertain to grain boundaries (or veins).

Finally, I feel that the topic of seeking to explain observed instances of excess diffusion (with bulk records) lies peripheral to the current study, to the subject defined by its title. To maintain a strong focus for the manuscript, I prefer not to extend it to treat or comment on the topic. The avenue of studying the pattern of excess diffusion alongside ice-core proxies has already been pointed in the Conclusions section of Ng (2023; p. 3079), which I think is a better place for the idea, because half of their paper is devoted to modelling the diffusion-rate depth profiles.

Minor points:

Line 40. A link is required. Done. Please see Line 38-39 in the uploaded manuscript.

Line 254. Change HCL to HCl

Done. Please see Line 237 in the uploaded manuscript.

I look forward to reading your response.

Sincerely,

Florent Domine

Minor changes applied to the manuscript:

20, word inserted: "times"

38-39, weblinks now given here

55, removed a comma (... excess diffusion potentially caused by...)

111, minor rephrasing to "models of the enhancement factor"

[and deletion of "based on the short-circuiting"]

170-171: Before the Thomson et al. (2013) reference, the vague phrase "under different conditions" has now been deleted. The phrase is redundant as their study is described later on Line 200-203. 188, changed "as" to "to be"

190-191, I removed some unnecessary definite articles here

194/195: I have swapped "latter" and "former". (In the last revision, these got mixed up when I constructed the text of this paragraph. This is a typographic mistake. The model has not changed.) 237: "HCL" corrected to "HCl"

493: I simplified this sentence in the caption of Fig. 4

525, typo corrected: "gradents" >> "gradients"

528, missing hyphen inserted

643-645: I edited this long sentence slightly to manage its items better.

672, rephrased "the dependence" to "this dependence"

724, added missing year "(2009)"

751: I changed "give" to "might give"

771, "weakest" changed to "weaker ... than elsewhere"

831, I spelled out "it" as "the section" for clarity

844, idea clarified by writing "fast HCl diffusion" instead of "fast diffusion"

866, word inserted: "times"