27 August 2025

Dear Editor Nanna Karlsson,

I write to submit our revised manuscript (egusphere-2025-1566) for your consideration. Please find here our point-by-point description of the revisions, alongside the reviews. We made all of the changes described in our replies to the reviewers (AC1 and AC2).

As promised, we have put our computed results in a FAIR repository, and now give the corresponding link in the Code and Data Availability statement.

Kind regards,
Felix Ng

Black: review comments. Blue: authors' response.
Red: revision change. The line numbers are those in the track-change-accepted manuscript.

RC1: Anonymous Referee #1

1 General
In this paper, the authors analyze the diffusion of key climate signals in ice cores, taking sulfate as the primary molecule of interest. They pick up the thread of the recent analyses by Fudge et al. (2024) and Rhodes et al. (2024), where the results of those papers differ, in part, due to their distinct methods for estimating the diffusivity. Felix Ng and company describe these differences and place their origin on a firmer theoretical foundation. The current authors then take the new theory inference and perform a new inversion. The results of this effort are sort of mixed and could be clearer (but maybe the answer is that the data / inversion is just inconclusively unclear). They then perform a forward simulation, which matches the data nicely and gives new insight. Then there is a discussion about the rate of diffusion through the firn. This part of paper is a little speculative and lacks some of the clarity from the earlier part of the paper.

Overall, this is a really well-written paper – Felix and team are articulate, poetic, and precise in their wording – a masterclass in writing. I like that this is an all-star team – with 3 recent papers on the subject – joining forces to get to the bottom of this question. I also like that this paper is eminently readable. There is a nice description of the physics of the $t - z$ transformation to $\psi - \zeta$. With a few minor changes, I am happy to support publication.

Thank you for your valuable review. It is very encouraging and reassuring for us to read your summary of our manuscript's thread and contributions. We are happy to know that our writing communicates the science well.

Regarding the part that considers diffusion through the firn (Sect. 4.1), in this manuscript we have space to consider the potential mechanism only at a simple level. We hope you agree that Sect. 4.1 foreshadows a full study; we plan to tackle that elsewhere.

2 Remarks

1. I think more discussion of DR is warranted – it turns out to be important and having not read Rhodes et al. (2024) in enough depth, I am wishing that it was clearer in the text.

From reading the comment and visiting the manuscript, it is not clear to us what is missing (and where). How the method of Rhodes et al. (2024) works and what D_R measures are explained on Lines 81–90, together with Fig. 1b. Their findings for D_R are reported later in the Introduction on Lines 105–122 (with Fig. 1c); there, the decay in D_R is described – positioned for us to return to address later in the manuscript. Sects. 2.2.1, 2.2.2, and 3.1 also refer to D_R in the context of our theory and results in various places. Presently we have not decided on revision on this matter. We think that the text strikes a reasonable balance in terms of explaining what our study needs and recounting enough of Rhodes et al. (2024), and we rely on readers looking up their referenced study for more background.

No editing changes made, as explained above.

3 Specific comments

1. Section 2.2.1 - it could be worth making the connection to the method of characteristics as motivation for the change of variables.

Thank you for this suggestion. We considered it but decide not to draw the connection. In this part of the manuscript, where we launch readers into the first heavy mathematical idea, it is better for us to focus on getting them safely to Eqs. (5) and (6) and the implications, without mentioning too much that is peripheral. We think that the concepts given on Lines 174 and 175 sufficiently signpost the motivations.

No changes made.

2. Line 192: what is the 4.2919 factor?

The factor converts the FWTM of the Gaussian to its standard deviation σ. We will extend Lines 192-193 to clarify this.

Done; see lines 192–193.

3. Lines around 320: is DE the same as Deff from before? I am missing the subtle difference.

DE is the effective diffusivity sought by the inversion approach based on mean absolute gradient, assuming D to be constant in the inversion interval, but not using the approximation made by Barnes et al. (2003) described on the last page (p12, lines 301–304). We will edit the writing near Line 320 to clarify the distinction.

Done; see lines 318–320.

4. Figure 4a: a legend or arrows could be helpful. It took some time for me to see the exponential curve.

Yes, we can add legends in Fig. 4a for the line types.

Done. Please see Fig. 4a. On lines 401 and 410, we also now write “two-term exponential” to coordinate with the figure legend.

5. Where is the first inversion approach plotted? Figure 5 is the second approach only, correct?

Figure 5 does plot the results of the first inversion approach, as described on Line 398: "In contrast, D(t) from the first approach (dashed black curve in all panels, Fig. 5) shows a much more…". Figure 5 also clarifies the origin of the curve in its caption and legends.

To help readers register this information, we have rephrased the underlined words above to

"In contrast, $D(t)$ retrieved by the first approach (Fig. 5, dashed black curve in all panels)…".

Please see line 400.

6. I am very confused by the oscillations in D(t) shown in figure 5. Is some sort of regularization required?

D(t) oscillates because the inversion finds stretches of negative D as well as positive D, so we have focussed on explaining how negative D can arise. Potential reasons include noise on the D_R values (which derive from estimates from the sulphate record), uncertainty around the amount of spline-smoothing, and time-dependent variations in the real system (e.g. history of ice submergence velocity in the column). We outlined some of these on Lines 430-432; in a revision, we will try to improve their wording and link in the observed oscillations. Although regularization might prevent or suppress the oscillations, we don't think it is the right way to go, given the causes mentioned above.

Done. We have improved the passage on lines 431–436 to clarify the causes behind the stretches of negative D and the oscillations.

RC2: Referee #2 (Anders Svensson)

The manuscript aims at quantifying the (effective) diffusivity of sulfate ions in the upper 2700m/~400ka of the EPICA EDC ice core. A nice theory for the involved processes is developed, at least for the upper part of the ice sheet, and based on results from previous work, the manuscript does come up an improved understanding of the topic and novel results. Overall, I mostly have a few comments to the manuscript, not any major concerns.

The manuscript is rather long, explaining every step taken in full detail. It would be an option to move some of the derivations into appendices. On the other hand, it is nice as a reader to be guided carefully through the various steps, so if it is not a concern of the journal, I would not recommend shortening it. In general, the manuscript is extremely well written and well illustrated. Obviously, a lot of effort has already gone into streamlining this manuscript.

The topic of determining the (effective) diffusivity at the EDC site has recently been the topic of two other papers in Climate of the Past. The current manuscript carefully explains why those studies reached deviating results/conclusions and elegantly develops a more advanced theoretical framework that is able to reproduce the previous results and explain where they went wrong/made too bold approximations. In particular, the diffusion of sulfate in the firn layer appears to be an important consideration that needs to be taken into account. The firn has been known to be important for diffusion of water isotopes for a long time, and it would be interesting to know which other components have important signal dampening by firn diffusion. And what about dust particles that are not supposed to diffuse importantly, can they teach us about what other processes are important for signal disturbance in the deeper ice?

Thank you for perusing our manuscript and your insightful review. Your comments on the broader science surrounding our study are much appreciated. They echo our thoughts and ongoing work beyond this study, e.g., some of the avenues outlined in the Discussion.

We are very happy to read your appraisal of our writing and your comments about its length. In a study that develops a new mathematical theory and applies it to reach new results as well as critiquing past methods, we feel the need to explain and report in detail.

Given our focus on sulphate and its diffusivity inversion, in the manuscript we have refrained from speculating/discussing how diffusion in the firn might impact other species or the role of dust on signal evolution. We agree that these topics are important for the field to address.

Still, with all the improvements introduced in the current manuscript it is remarkable to see in Figure 9 that all of the estimated contributions to the sulfate signal diffusivity are INCREASING with depth/age whereas the observed diffusivity clearly is decreasing with depth/age even with a temperature profile that increases with depth. Indeed, there are additional layers of understanding of the topic to disentangle in future studies. Clearly, a lot can be explained be letting the different suggested types of diffusion phase in at out at different depths/ages, but as I see it, there must still be something quite fundamental lacking for our understanding of the topic. For example, the effect of dust or other impurities, the development of size and/or orientation of ice crystals with depth. Let alone, the strange-looking sulfate peak shapes observed in the deeper sections of the EDC ice core (not something that has been observed in Greenland to my knowledge). Hopefully, soon-to-appear very high resolution records of ion locations and other properties in ice cores will enlighten us.

In a revision, we could inject the idea of possible additional factors regulating the diffusivity profile into Sect. 4.2 or 5. We shall keep this very brief, aiming mainly to sketch the challenges ahead and make our text more rounded.
Done. Please see lines 825 – 827, where we added the pointer:

"There is also possibility for signal modification by mechanisms involving more than just diffusion, e.g., reactions between different compounds or between dissolved ions and dust particles."

The reviewer's idea of ice fabric/textural changes already features in a different guise near the end of Sect. 4.2 (lines 736-737), where we highlight:

"potential anisotropy in grain-boundary motion and orientation (which may impact *D*res)" as one of the limitations behind our interpretation of the retrieved diffusivity profile.

"As for the new cores and high-resolution records, we too look forward to learning what they teach us!

Although the study is concerned with sulfate spikes in the EDC ice core only, the developed theory can be applied to other ice cores provided high-resolution sulfate records and other essential records are available. As I understand it, the theory could even be applied for other types of ions or impurities provided the necessary records are available. The EDC core is of particular interest due to the recent arrival of the BE-OIC oldest continuous ice core from Antarctica that currently has a lot of attention and for which the preservation of signal strength in the deepest ice is a major point of interest. A logical next step for the present study would be to apply the developed theory to other ice cores and to get a more general overview of the signal preservation of impurities in ice with different properties. For example, it would be very useful to know which magnitude of volcanic eruptions that will survive to which depth/age in different ice cores, eg a figure 10 that covers other ice cores. It is understandable, however, that the current manuscript does not cover those topics.

Thanks for these helpful comments. The theory can be applied to other ions/impurities and other cores (as mentioned on Line 60). With the EDC core, nitrate and chloride may be the more obvious ions to study. Our immediate next focus, however, is on other cores, as the finding of high sulphate diffusivity in the firn intrigues us.

We read your idea of repeating Fig. 10 for other cores with enthusiasm, although we can see that estimating signal survival at their sites will require much bolder assumptions (about the vertical pattern of diffusivity) than those we currently use in Sect. 4.3 for the Dome C region. On lines 807-808, we added a passage to indicate what would be required to compute the equivalent of Fig. 10 for other cores.