

Reply to Reviewer 1, Alan Rempel.

We are grateful to Alan Rempel for his constructive and thoughtful review of our manuscript. We are confident that all concerns raised can be addressed in a revised manuscript. The Discussion section will be improved thanks to his insights on the physical processes involved.

We do not reproduce the entire review here – sections of it are shown in italics. Our replies are given in regular font.

The discussion of previous theoretical results in the introduction is appropriately concise, but might be altered slightly to some benefit. On line 45, the 50 cm displacement noted in the abstract of the paper by Rempel et al. (2001) is quoted without sufficient context to enable the reader to understand the conditions that led to that particular figure, which applied to ice of Eemian age in the GRIP ice core. Since that theory would predict different displacements in the EPICA Dome C core at different depths, I'd suggest rewording to something like: "The implication was that a chemical signal of Eemian age in the GRIP ice core ...".

This suggestion will be adopted in a revised manuscript.

Regarding the following sentence, a half meter doesn't seem like much in a >2km deep core, so it isn't clear whether the consequences for cross-matching events between ice cores for stratigraphic purposes would in fact be "major", or typically quite minor { perhaps the adjective should be removed.

50 cm is indeed a small interval of depth but at the base of an ice core, where layers are thinned, it equates to a substantial amount of time. The statement will be qualified in a revised manuscript.

On line 50, the claim is made that Ng's (2021) modified theory would prevent such compositional displacement, but destroy the chemical signals over time. This would be somewhat unsatisfying given that chemical signals clearly persist for long durations, but Ng does show that deep signals can remain intact if the effective diffusivity is reduced or if spatial variations in grain size are invoked. I'd suggest appending the sentence with something like: "... they will be destroyed over time if they are free to diffuse unimpeded through connected veins into adjacent low concentration regions."

This suggestion will be adopted in a revised manuscript.

The differences between the Rempel and Ng treatments are not central to this manuscript, but as the former model is disregarded following the opening sentences of this paragraph, for further context I think it worth clarifying my own understanding of the primary difference between these formulations...This has been a rather verbose digression. Perhaps rather than saying that the Gibbs-Thomson effect was neglected, it would be more correct to say something along the lines of: "... challenged the impact of this phenomenon by noting that since soluble impurity content appears not to exert a dominant control on ice grain size (e.g. Durand et al., 2006) and by extension, vein density, the Gibbs-Thomson effect should cause vein radii to adjust by producing solute concentration gradients that diminish bulk concentration anomalies."

We thank the Reviewer for taking the time to detail his insightful thoughts. We will certainly consider them carefully and likely adopt the modification suggested in a revised manuscript.

The approximately Gaussian form of observed volcanic sulfate anomalies is somewhat curious (line 74). One might have expected fallout and deposition to be concentrated at first and subsequently diminish over time and so be "front-loaded" to some extent. Based on modern observations, could you comment on whether the Gaussian shape results from short-term post-depositional changes (e.g. due to Gibbs-Thomson diffusion), or whether this is instead the characteristic pattern of volcanic fallout from stratospheric levels?

The reviewer is of course correct that the accumulation of sulfur in the stratosphere and its subsequent deposition to the ice sheet will be asymmetric, with a relatively sharp onset and a decaying tail. This asymmetry is indeed seen in recent eruptions, but observations of eruption signals at EDC shows that the signals tend towards symmetry within a time (of order 1 kyr) that is relatively short compared to the >400 kyr of this study. For this reason, we chose the mathematical simplicity of assuming a Gaussian shape from the start. Our sensitivity studies (Fig. 3) where we changed the width of the initial peak between 1 and 5 years show that the exact width or shape of the initial peak does not materially affect the derived effective diffusion coefficients.

In Figure 1, comparisons of the displayed scale bars showing 5 yr of ice accumulation with the observed sulfate peaks provide vivid illustrations of post-depositional effects. However, the 5 yr span collapses onto a vertical line in the final 3 examples. I appreciate that the text gives further context, with the quoted 30 yr span for the 364 ka peak. However, I'd suggest modifying the figure caption or annotating each panel with the number of years that the 1m depth range represents.

This suggestion will be adopted in a revised manuscript.

I found the theoretical development in sections 3.2 and 3.3 somewhat confusing. The standard convention in the modeling literature with which I am most familiar is to treat equations as portions of sentences, with appropriate punctuation (e.g. see Ng's 2021 paper). Instead, here you refer to the equations by number, and subsequently separate them out from the text. To me, this seems disjointed.

We agree with the Reviewer's opinion and will modify sections 3.2 and 3.3 accordingly.

In equations (6) and (7) you note that the effective diffusivity is expected to be a function of time. However, my understanding is that your model calculations in fact treat the diffusivity as constant through time is this correct?

Yes, the Reviewer is correct. Effective diffusivity is treated as constant for each volcanic peak – it does not evolve with time. Equations 6 and 7 will be modified in a revised manuscript, following also the advice of Reviewer 2.

It wasn't immediately obvious to me how equation (12) came about and why there is no explicit dependence on time. Indeed, a is really a rate, so I believe at/H is needed in the argument of the exponential to ensure dimensional correctness, and integration of (9) would produce this result following correction of a sign error.

Yes, the equation should indeed have at/H in the exponential.

On line 248 temperature and chemistry are mentioned as controlling variables. Perhaps grain size, or more generally, microstructure, should be mentioned as well.

This suggestion will be adopted in a revised manuscript.

On line 295 the very low eutectic temperature of sulfuric acid is used to justify the expectation that sulfate ions can be dissolved in liquid at EDC temperatures. However, in the paragraph beginning on line 155 you mention the Traversi et al. (2009) finding that appears to suggest that sulfate reacts with dust to presumably form a solid precipitate. Would it possibly be worth saying more here about the potential effects of chemical reactions between different impurity species?

This suggestion will be adopted in a revised manuscript.

The brief discussion in 5.3 begins by noting that the simplest version of Ng's (2021) model predicts much faster diffusion than is observed in the Holocene ice, which itself is faster than that observed in deeper regions. That the inferred diffusivity does not appear to depend on signal size would also seem to differ from Ng's (2021) model predictions. The proffered suggestion that Gibbs-Thomson diffusion efficiently reduces vein concentration gradients would appear to effectively transform Ng's model to the Rempel et al. (2001) model, albeit only if vein density can evolve to enable signal translation. As the Barnes et al (2003) treatment relies upon effects of grain-size evolution, it perhaps might contain some of the essential elements that these other two models lack. I'm not sure I follow the reasoning behind the final sentence of this section. You've shown that the effective diffusion rate in the Holocene and early Pleistocene is both much slower than Ng's Gibbs-Thomson mechanism would predict and not systematically dependent on anomaly magnitude, so what makes you conclude that Ng's model correctly describes the controlling mechanism? I thought that I understood the Barnes-type model to depend on grain growth, but in the final clause you say that the rate of grain growth isn't important. Please clarify.

We thank the Reviewer for these insights thoughts and queries. They were certainly help us to clarify our arguments when modifying Section 5.3.

There's a typo in the title of the penultimate reference.

Thanks.