

Interactive comment on “Methanesulfonic acid (MSA) migration in polar ice: Data synthesis and theory” by Matthew Osman et al.

B. Alexander (Editor)

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Dear Dr. Alexander,

We thank you for your suggested technical corrections, which are addressed below. For convenience, your comments are reproduced below, our replies to them are in italics, and excerpts from the text are between double quotes.

Dear Matthew Osman,

Thank you for submitting your responses and revisions. In addition to the reviewer comments that you have already addressed, I have a few minor suggestions detailed below that I think will improve clarity. Please let me know if you have any questions.

Regards,

Becky Alexander

Page and line numbers below refer to the revised manuscript.

Suggested technical corrections

C1) Page 2 Line 14: Photolysis also results in postdepositional alteration of some chemical species. Perhaps this should be explicitly mentioned in this general statement about possible processes impacting chemical species in ice cores.

We have now included “photolysis” in the opening paragraph of the manuscript (Pg. 2, Line 14):

“Processes acting within the upper firn layer, including wind pumping, diffusion, photolysis, volatility, sublimation, and melt (Wolff et al., 1996), can affect the stability of chemical species... (.)”

C2) Page 3 Line 9: “lack of wintertime MSA deposition” implies zero MSA deposition flux during winter. Do we really know that it is zero? MSA has an atmospheric lifetime of several days, and thus can be transported over significant distances before deposition. Perhaps replace “lack of” with “relatively low”?

We agree with this suggestion, and have replaced “lack of” with “relatively low” (Pg. 3, Line 9):

“Conversely, the relatively low wintertime MSA deposition may be jointly attributed to... (.)”

C3) Page 3 line 11: “OH-“ should read “OH \dot{E} S’ ” or “OH radical”.

We have changed “OH” to “OH radical” on Pg., 3, line 12.

C4) Page 6 lines 19 and 24 have confusing wording. “increasingly shallow depths” is confusing. Maybe replace with a description of the relationship between snow accumulation rate and the depth over which MSA migration occurs. “depths lower” is also confusing. Does “lower” mean above or below a certain depth in this context?

We have removed “increasingly shallow depths” on Pg. 6, line 19, and have reworded the sentence as follows (Pg. 6):

“At low-to-moderate accumulation rate sites ($\dot{b} = 0.1 - 0.45 \text{ m w. eq. yr}^{-1}$), MSA migration seems to universally occur, with the shallowest reported depths of migration showing a positive relationship with accumulation rate (Table 1 and Fig. 2).”

On Pg. 6, Line 24, we have changed “depths lower” to “depths deeper”.

C5) Page 9 line 7: Does this mean that the depth at which movement of MSA stops is deeper, or that MSA moves a larger distance in total?

On Pg. 9, Line 7, we make the observation that the shallowest observed depth of MSA migration (z_{fo}) appears to occur at deeper depths for sites with lower core-averaged Na^+ concentrations ($\overline{\text{Na}^+}$). This observation does not necessarily relate to the distance traveled by MSA from a summer layer to a winter layer. The sentence referred to on Pg. 9, Line 7 has been adjusted to read as follows:

“We find that, as $\overline{\text{Na}^+}$ decreases, MSA migration tends to be observed at greater depths in the firn or ice column (Fig. 3).”

C6) Page 9 line 8: Can “small values” be replaced with “low concentrations”?

We have changed “small values” with “low concentrations” (Pg. 9, Line 8).

C7) Page 20 line 7: The textbook Seinfeld and Pandis [2006] is probably not the appropriate reference for the Cl:Na molar ratio in seawater.

The reference for Seinfeld and Pandis (2006) has been replaced with that of Chesselet et al. (1972; see references below), who similarly adopted a molar ratio of Cl:Na = 1.8 for seawater to study variations in ionic ratios of marine aerosols.

C8) Page 32 line 6: Can you be more quantitative with the statement “high accumulation and low core-averaged $[\text{Na}^+]$ ” by giving some numbers of range of numbers for “high” and “low”?

Our results suggest that the timing/depth of MSA migration is a continuous function of site accumulation rate (\dot{b} ; positive relationship) and core-averaged $[\text{Na}^+]$ ($\overline{\text{Na}^+}$; negative relationship), indicating that migration should inevitably occur provided i) enough time and (or) depth and ii) requisite thermal conditions (Sect. 5.1). As such, a conscientious decision was made in refraining from providing a specific range of \dot{b} and $\overline{\text{Na}^+}$ values for which MSA migration may be deemed “negligible”. We provide two primary reasons in support of this decision:

- 1. Studies using [MSA] for paleoclimatic inferences vary in terms of timescales and temporal resolutions that are explored/achieved (Table 1), indicating that values of \dot{b} and $\overline{\text{Na}^+}$ which are relevant will similarly vary from study to study.*
- 2. Following reason (1), irrespective of the site-specific values of \dot{b} and $\overline{\text{Na}^+}$, the precise timing of MSA migration still remains broadly unconstrained, given in particular the significant uncertainty in the grain-boundary diffusion coefficient of MSA (Sect's. 4.4; see also Fig. 14).*

Thus, providing quantitative constraints on “high accumulation and low core-averaged $[\text{Na}^+]$ ” may be subjective and (or) potentially misleading. Rather, we believe the associated quantitative results are well encapsulated within Figures 14 and 15. As such, no changes were made to Pg. 32, Line 6.

References

*Chesselet, R., Morelli, J., and Buat-Menard, P.: Variations in ionic ratios between reference sea water and marine aerosols, *J. Geophys. Res.*, 77(27), 5116–5131, doi:10.1029/JC077i027p05116, 1972.*