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Post-depositional migration and signal reconstruction of methanesulfonic acid (MSA) in polar ice cores

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Methanesulfonic acid (MSA; CH_3SO_3H) in polar ice cores is a unique proxy of marine primary productivity, synoptic atmospheric transport, and regional sea ice behavior. However, MSA can be unstable within the ice column, leading to uncertainties surrounding the integrity of its paleoclimatic signal. Here, we use ice core records coupled with forward and inverse numerical models to investigate the post-depositional processes affecting the migration of MSA within the firn and ice column, and attempt to reconstruct the original signal in the ice column. The forward model, detailing the vertical diffusive transport of soluble impurities through supercooled liquid pathways, allows us to systematically assess the contribution of varying influences on the post-depositional migration of MSA. Our results show that two site-specific variables in particular, i) snow accumulation rate, and ii) seasonal concentration gradients of Na^+(typically the highest concentration sea salt), may be sufficient to reasonably predict the timing and magnitude of MSA migration within the ice column. However, at present the temporal accuracy of the forward MSA migration model remains limited by inadequate constraints on the diffusion coefficient of MSA, D_{MS-}. Specifically, we find that previous estimates of $D_{MS-}$are unable to reproduce, within significant uncertainty, the progressive phase alignment of the MSA and Na^+signals observed in real Antarctic ice cores.

To attempt to correct for the effects of post-depositional migration, we combine recent high-resolution West Antarctic MSA data using sequential methods from optimal control theory (a Kalman filter and a related fixed-interval smoother) to reconstruct and provide uncertainty estimates on the original, pre-migrated MSA profile. We find that although the reconstructed MSA profile provides a reasonable estimate of the original MSA signal, the large uncertainties associated with this reconstructed signal cannot be objectively discriminated against the migrated MSA data. On the other hand, our results suggest that records undergoing severe migration may still be useful for inferring $\sim$decadal and lower-frequency climate variability.