

Interactive comment on “The sensitivity of the oxygen isotopes of ice core sulfate to changing oxidant concentrations since the preindustrial” by E. D. Sofen et al.

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We thank the reviewer for providing helpful comments on the manuscript. Specific responses to the reviewer's comments are detailed below.

Introduction seems a bit thin and could improve a bit in defining the existing problems and challenges the paper will explore.

The introduction (Sections 1 and 2) has been extended with more background information, a statement of the purpose of the paper, and a outline of the structure of the paper.

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It appears that one can discuss, in the introduction, that D17O record is not straightforward because despite of O3 increase from PI to PD, D17O decreased in both Greenland and Antarctica from PI to PD. This is opposite from what one expects for a linear response to O3 level.

Thanks to the reviewer for suggesting this clear way of presenting the motivation for this study. Reference to this are included in the abstract and in section 5.

The paper concludes that the decrease in pH and increase in metal catalyst counteract increased O3 level. From this observation alone, it appears that the D17O of sulfate is probably the most sensitive to metal catalyst loading or pH (NOx and SO4²⁻) of the rainwater than oxidant levels. I am slightly confused with the logic in the abstract. “The modeled D17O-SO4 is consistent with measurements. . .” Was the model fit to the observation using free parameters (i.e., pH and metals?) to generate consistent D17O values?

The discussion of pH in Sections 4 and 5 has been extended. In those sections, we elaborate upon the choice of cloud water pH values. While the pH values chosen for use in the sensitivity studies do represent the best-fit cases, they are also justifiable on the basis of other observational evidence (measurements of cloud water, precipitation, and ice core pH). In addition, although the simulated $\Delta^{17}\text{O}_{\text{SO}_4}$ in each scenario is highly sensitive to pH, the PD-PI change in the $\Delta^{17}\text{O}_{\text{SO}_4}$ is consistent for each pH (4.5, 5.0, 5.5) simulation at constant pH (constant pH can be assumed for the Southern Hemisphere). Metals were not varied to match $\Delta^{17}\text{O}_{\text{SO}_4}$, but are dependent upon anthropogenic emissions and dust, as described in detail in Alexander et al. (2009).

The last sentence in the abstract may need more explanation.

Changed to “uncertainties in the preindustrial emissions of oxidant precursors.” rather than uncertainty in the oxidants themselves, as we vary emissions from biomass burning and emissions of NO_x and VOCs.

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The manuscript does not clearly state sensitivity of D17O sulfate for oxidant levels as title suggests. It appears it would be a very powerful proxy if one could constrain the rainwater pH.

As described above, by focusing on the $\Delta^{17}\text{O}_{\text{SO}_4}$ difference between PD and PI simulations, the uncertainty due to pH can be mitigated on this timescale. However, it remains to be seen whether the pH issue can be overcome on other timescales over which pH has varied substantially (e.g. glacial-interglacial). We show that Greenland $\Delta^{17}\text{O}_{\text{SO}_4}$ is not sensitive to oxidant variability in the northern hemisphere due to the dominant controls of metals and pH. Since variability in metal concentrations and pH is negligible in the SH, Antarctic $\Delta^{17}\text{O}_{\text{SO}_4}$ is sensitive to oxidant variability. However, we demonstrate that the oxidants have changed in such a way to have no net effect on $\Delta^{17}\text{O}_{\text{SO}_4}$. Please see section 5 for a revised discussion of this.

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