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Title: "Reactions of Carbonyl Oxide with Aldehydes: Accurate Electronic Structure Methods, Kinetic Insights, and Atmospheric Implications"

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Responses in blue.

Editor:

Please correct the sorting of your figures: figure 1 is currently appearing after figure 4.

Author response: Thanks for your comments. We have corrected the placement of the figures in the revised manuscript. Figure 1 has been moved to its correct position.

RC1

I have only one very small comment left: based on comparing figures 5c and S10, it seems like the large percentage changes in gas-phase H₂SO₄ (called "gas phase sulphate" in the paper for some reason) all occur in sections where the absolute concentration is rather low. And vice versa, in the regions where the absolute concentration is high (e.g. the 1E8 value quoted in the text for Mexico), the percentage change is negligible. This ought to be mentioned in the manuscript.

Author response: Thanks for your comments. regarding the terminology, we would like to respectfully clarify that "gas-phase sulphate" is indeed the intended term and not a typographical error for gas-phase H₂SO₄, it specifically refers to the SO₄ radical in the gas phase in the GEOS-Chem model. we completely agree with your assessment regarding the spatial distribution of the percentage changes. The large relative changes are mathematically driven by the low baseline absolute concentrations in those specific regions. Conversely, in regions with high absolute concentrations (such as the 1×10^8 value over Mexico), the relative changes are naturally negligible. This is a crucial point for correctly interpreting the data, and we appreciate the suggestion to make it explicit. We have added the discussion in the revised version "While these relative changes might initially suggest a significant regional sink for atmospheric sulfate aerosols, based on a detailed comparison of Figures 5c and S10, we indeed find that the largest percentage changes in gas-phase H₂SO₄ predominantly occur in regions with relatively low baseline concentrations. Specifically, while peak concentrations over Canada and Russia can reach 10^7 molecules cm⁻³, their regional averages remain around 10^5 molecules cm⁻³. In contrast, in regions with high absolute concentrations (e.g., $\sim 1 \times 10^8$ molecules cm⁻³ over Mexico), the relative changes are minimal. This reflects the fact that even modest absolute variations can translate into large percentage changes when baseline levels are low, whereas similar or larger absolute changes appear less significant in high-concentration regions. Consequently, this reaction ultimately exerts a negligible effect on the overall global reduction of atmospheric sulfate." in page 27

RC2:

The authors have provided satisfactory changes and/or responses to most of the issues I raised with the original version of the manuscript. The only remaining concern is with Figure 5. There should be an explicit discussion of the right-most panel, which shows

changes in global sulfate concentration. Presumably, the reductions of nighttime sulfate over Canada (12.2%) and Russia (6.01%) presented in the second-to-last sentence of Section 3 are depicted in Figure 5. The authors should cite Figure 5 there.

Author response: Thanks for your comments. We completely agree that the right-most panel of Figure 5, which illustrates the spatial distribution of these changes, should be explicitly cited and discussed in the text to better support our quantitative findings. We have added the discussion in the revised version “While the relative changes might initially imply a substantial regional sink for atmospheric sulfate aerosols, a detailed comparison of Figures 5c and S10 reveals that the largest percentage changes in gas-phase sulfate predominantly occur in regions with low baseline concentrations. Specifically, although peak concentrations over Canada and Russia reach $\sim 10^7$ molecules cm^{-3} , their regional averages remain on the order of 10^5 molecules cm^{-3} . In contrast, regions with much higher absolute concentrations (e.g., $\sim 1 \times 10^8$ molecules cm^{-3} over Mexico) exhibit only minimal relative changes. This indicates that modest absolute variations can produce large percentage changes under low-background conditions, whereas comparable or even larger absolute changes appear insignificant in high-concentration environments. Consequently, this reaction has a negligible impact on the global atmospheric sulfate burden.” in page 27