Manuscript No.: ACP-2026-68
Title: In Situ Real-Time Determination of SO2 Photochemical Oxidation in Nanoscale Sea Salt Aerosols based on Dark-Field Microscopy

We thank the anonymous referees for their valuable and constructive comments/suggestions on our manuscript. We will revise the manuscript accordingly and please find our point-to-point responses below.

Comments by Anonymous Referee #2:

General Comments:

This manuscript details a method using dark-field microscopy for in situ real-time measurement of photochemical oxidation of SO_2 in individual nanoscale aerosol particles. Hygroscopic growth factors and the Zdanovskii-Stokes-Robinson rule are employed to determine first-order kinetics for sodium chloride aerosols ranging in size from 50-400 nm, revealing a non-monotonic dependence of the reaction rate constant on particle size. Additionally, the study investigates the effect of organic coatings on oxidation reaction rates. The method is certainly novel and an appropriate topic for ACP, however I have several questions and points of confusion that should be addressed prior to acceptance.

Response: We thank the reviewer for the constructive comments and for recognizing the novelty and relevance of this work. We also appreciate the reviewer's careful reading and for pointing out several questions and areas that require clarification. Following these comments, we will revise the manuscript to improve the clarity of the data analysis, better explain the experimental design and underlying assumptions, and strengthen the presentation of the results and their interpretation. Point-by-point responses to the reviewer's comments are provided below, and all corresponding revisions will be incorporated into the manuscript.

Major comments:

1) *I found it difficult to follow the method for calculating the rate constant as detailed in the experimental section, and then subsequently challenging to connect the presentation of kinetic data in the results with the described calculations. There were several mislabeled variables (see Minor Comments for examples) or new variables introduced in the discussion that were not*

defined in the method (specifically $ln(C_0/C)$. Most of my confusion re: calculations will be alleviated by correcting equation/variable labels and clarifying how plotted variables in Figures 3-5 were determined, but as it currently is presented, the method for data analysis is unclear and needs to be addressed.

Response: We thank the reviewer for this careful reading and for pointing out the lack of clarity in the presentation of the data analysis procedure. In response, we will revise the relevant sections of the manuscript to improve the transparency and consistency of the kinetic analysis.

Specifically, the calculation procedure for deriving the rate constant will be reorganized and described in a clearer, step-by-step manner. All variables used in the analysis (including $ln(C_0/C)$) will be explicitly defined upon first introduction, and their physical meanings will be clarified. In addition, we will carefully check and correct mislabeled variables and ensure consistency between the Methods section and the presentation of results in Figures 3 – 5.

Furthermore, we will expand the description of how the plotted quantities were obtained from the experimental measurements, including the derivation from hygroscopic GF data to the kinetic parameters. These revisions are intended to eliminate ambiguity and ensure that the data processing workflow is transparent and reproducible.

2) *The primary conclusions regarding the non-monotonic dependence of the SO_2 reaction rate constant on particle size rely on a comparison of the experimental GFs of NaCl particles with E-AIM calculated GFs of NaCl and $NaHSO_4$ particles. Comparison to experimental GFs for NaCl and $NaHSO_4$ particles or literature GFs under similar environmental parameters would lend greater validity and confidence to the results presented in this work.*

Response: We thank the reviewer for this valuable suggestion. To strengthen the validity of the GF-based analysis, we will supplement the manuscript with experimentally measured hygroscopic GFs for both NaCl and $NaHSO_4$ particles under comparable environmental conditions. The experimental results will be included and compared with the corresponding E-AIM model predictions.

3) *How might this method be applied to cover a scope of more atmospherically relevant conditions, such as lower SO_2 concentrations or lower RH?*

Response: We thank the reviewer for this important question regarding the broader applicability of the method. We acknowledge that atmospheric conditions often involve lower SO_2 concentrations and a wider range of RH. In principle, the methodology developed here can be extended to such conditions.

The present study focuses on the retrieval of reaction kinetics and the investigation of curvature effects. To better resolve these effects and enhance the observability of the reaction behavior, relatively elevated SO_2 concentrations and high RH conditions were employed.

Future work will extend this approach to the investigation of more atmospherically relevant particles under realistic environmental conditions. We will clarify these points in the revised manuscript and identify this as an important direction for future work.

Minor Comments:

1) Figure 1: Misspelling of 'diffusion dryer' in graphic labels.

2) Line 100: The text refers to Figure 2.1 – should it be Figure 1?

3) Line 115: Please define the acronym 'OVF'.

4) Line 150: Should the section title read '2.4 Calculation of Reaction Rate'?

5) Line 178: Should the defined variables be C0, Ct? I do not see CT in either equation (7) or (8).

6) Figure 2/Line 210: The text refers to a red curve representing fresh NaCl particles in Figure 2, but the curve in reference is actually blue?

7) Lines 240-250: The text refers to ln(C/C0) but the corresponding Figure 3 (and later discussion) refers to ln(C0/C).

8) Line 350: The text referring to Figure 5 discusses ln(C0/C) values, but they are not represented in the figure?

9) Figure 5: The figure caption refers to a shaded area denote 95% confidence intervals, but there does not appear to be a shaded area in the figure.

Response: We thank the reviewer for the careful reading of the manuscript and for pointing out these minor errors and inconsistencies. We have carefully revised the manuscript to correct all typographical errors, inconsistencies in figure labeling, and unclear definitions. Specific revisions are detailed below:

1) The spelling of "diffusion dryer" in Figure 1 will be corrected.

2) The figure reference in Line 100 will be corrected to the appropriate figure.

3) The acronym "OVF" (organic volume fraction) will be defined upon first use in the revised manuscript.

4) The section title will be corrected to “2.4 Calculation of Reaction Rate.”

5) The variable notation will be corrected for consistency, and the definitions of C0 and Ct are now clearly stated in both the text and equations.

6) The color description of the curve in Figure 2 will be corrected to match the actual figure.

7) The notation for the logarithmic expression will be corrected to ensure consistency between the text and figures (ln(C0/C)).

8) The description of Figure 5 will be revised to accurately reflect the data presented, and the corresponding variables are now clearly indicated.

9) The figure caption will be revised to clarify that the shaded regions representing the 95% confidence intervals correspond specifically to Figures 5c and 5d.