

General Comments:

This manuscript details a method using dark-field microscopy for in situ real-time measurement of photochemical oxidation of SO₂ 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.

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.
- 2) The primary conclusions regarding the non-monotonic dependence of the SO₂ 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 NaHSO₄ particles. Comparison to experimental GFs for NaCl and NaHSO₄ particles or literature GFs under similar environmental parameters would lend greater validity and confidence to the results presented in this work.
- 3) How might this method be applied to cover a scope of more atmospherically relevant conditions, such as lower SO₂ concentrations or lower RH?

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 C_0 , C_t ? I do not see C_T 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 $\ln(C/C_0)$ but the corresponding Figure 3 (and later discussion) refers to $\ln(C_0/C)$.
- 8) Line 350: The text referring to Figure 5 discusses $\ln(C_0/C)$ 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.