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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Multiphase Reaction of SO_2 with NO_2 on $CaCO_3$ Particles. 1. Oxidation of SO_2 by NO_2" *by* Defeng Zhao et al.

Anonymous Referee #2

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This study investigated the heterogeneous reaction of SO2 with NO2 on individual CaCO3 particles in N2 using Micro-Raman spectroscopy. The results show that CaCO3 was first converted to Ca(NO3)2 forming a droplet and promoting the oxidation of SO2 by NO2.The precipitation of CaSO4 was suggested as a key step accelerating the sulfate formation. Based on the uptake coefficient determined, the authors concluded that the SO2 + NO2 reaction was not important compared to the oxidation of SO2 by OH radicals. The experiment was well designed and the paper was well written. But I do have concerns about the role of CaSO4 precipitation and I would also suggest the authors to compare their results with literature data before making strong statement on the role of NO2+SO2 chemistry.

Major concern:

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Discussion paper

1. The authors generalized the results of their CaCO3 experiments to assess the role of NO2+SO2 chemistry. I am not sure if such generalization is correct because according to early studies of Lee and Schwartz,1983 and Clifton et al.,1988, this reaction can be important under polluted and less acidic conditions in contrary to the authors' statement. The authors used deposited super-micro particles in their experiments. But I don't expect much difference between such a system and bulk experiments because large particles are not subject to strong Kelvin effect and particles contacted with substrates would not become supersaturated solution of high ionic strength due to nucleation. Thus before generalizing results for ambient aerosols, I would suggest the authors to discuss their difference with those early studies.

2. Based on Equation (5), the authors concluded that the precipitation-induced reduction of sulfate will promote the oxidation of SO2 by NO2 (reaction 2). I don't know if it is correct to use Eq. (5) in this way. Because Equation (5) is valid for reversible reactions and removing/adding products of non-reversible reactions will not change the reaction rate much.

Other comments:

Page 5 line 133, half sentene?

Page 6 line 187, I would suggest to briefly describe the mechanism of Ca(NO3)2 formation. Will the present of SO2 influence the uptake of NO2?

Fig. 4, no data for nitrate and carbonate after 120 min, why?

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