

Reviewer #2

Comment #1: The manuscript of Qiu et al., reported the influence of chloride chemistry to particulate nitrate formation in the framework of CMAQ. The authors included several chloride reactions into the CMAQ chemical mechanism. This topic is in the scope of ACP, and would benefit the knowledge of the heterogeneous reactions in the formation of particulate nitrate in Beijing, China. While in this paper, the detail of the parameters lacking the foundation and the analysis of model result seems common. Some of the conclusions seems can't obtain from the model result analysis (see major comments). The following comments should be addressed before publishing in ACP.

Response #1: Thanks for the positive comments on this manuscript. The suggestions are addressed in detail in the following.

Comment #2: Line 250-252, the treatment of aerosol surface area by time 5 or 10 in this model is unconvinced. Should provide more evidence to support the rationality.

Response #2: Figure R1a below is the detailed comparison of our modeled surface area with observations averaged between June 11-15, 2017. Under-prediction of surface area varies between 6-12 times. CMAQ model performance for surface area has not been extensively studied. In the only study we found in the literature (Park et al., 2006), similar under-prediction of particle surface area was reported. In their study, the mass concentration of PM_{2.5} at Atlanta, United State is generally well reproduced by the CMAQ model but the surface area shows large under-predictions similar to what we have seen in our study (see Figure R1b).

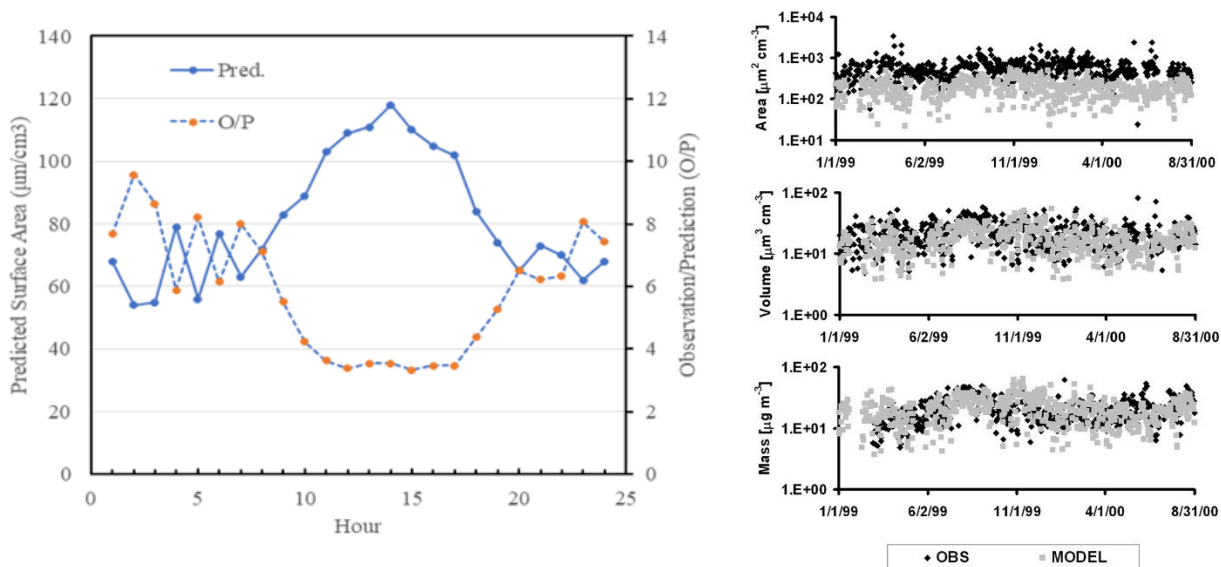


Figure R1a (left panel): Predicted averaged hourly particle wet surface area at IAP (June 11-15, 2017) and the ratio of observed to predicted PSA (O/P). R1b (right panel): CMAQ predicted and observed daily particle surface area, volume, and mass concentrations for PM_{2.5} from 1/1/1999 to 8/31/2000 at the JST station in Atlanta, Georgia. (R1b is adapted from Park et al., 2006).

Reference:

Park, S.K., Marmur, A., Kim, S.B., et al.: Evaluation of fine particle number concentrations in CMAQ. *Aerosol Science and Technology*, 40, 985-996, 2006.

Comment #3: The sensitivity tests used an O₃ uptake coefficient enlarged by a factor of 10 without any reference, while the Cl₂ simulations do not significantly improved in general. Other possible Cl₂ formation channel shall be tested or at least discussed.

Response #3: In Keene et al. (1990) [It was incorrectly cited in the original manuscript as Keene et al. (1999). We apologize for this mistake.], the daytime uptake coefficient of O₃ was not based on direct experimental measurements but was estimated indirectly based on a steady-state analysis of Cl₂ production rate in a hypothesized geochemical cycle of reactive inorganic chlorine in the marine boundary layer using a 0-D box model. Such an analysis tends to have large uncertainties and Keene et al. (1990) proposed 10⁻⁴ to 10⁻³. Nighttime value of 10⁻⁵ was proposed without much supporting evidence. We chose to increase these uptake coefficients to explore the upper limit of the impact of O₃ uptake on Cl₂ formation. As there are additional Cl₂ formation pathways, our results show that the higher uptake coefficients alone do not lead to significantly higher Cl₂ concentrations. The other Cl₂ channels are summarized in Table 2 and contributions of these channels to Cl₂ concentrations are explored in a separate study as this paper is focused on nitrate.

Comment #4: Line 398-402, I cannot agree with that the parameterization method including chloride of the uptake coefficient of N₂O₅ has a better performance, at least this kind of conclusion cannot be deduced from the authors analysis (cf. figure 1).

Response #4: Agree. This is removed from the conclusion section.

Comment #5: I suggest that the authors may present a table to summarize all the revisions of the parameters related to the uptake coefficient as well as the related heterogeneous reactions.

Response #5: We summary the revision of parameters in Table S3.

Comment #6: Line 206, Bertram et al., 2009 correct to Bertram and Thornton, (2009).

Response #6: Revised

Comment #7: Line 335-line 340, this part is confused. Did you mean more N₂O₅ convert to nitrate due to the N₂O₅ uptake coefficient calculated by Bertram and Thornton, (2009) is higher than the base case? And the non-significant nitrate increase may be due to the ClNO₂ yield buffered the increasing caused by the application of new N₂O₅ uptake coefficient?

Response #7: This sentence is revised as ‘By incorporating the chlorine heterogeneous reaction, the N₂O₅ concentrations decrease by about 16% because the uptake coefficient calculated with the Bertram and Thornton (2009) is higher than that with Davis et al. (2008).’

Comment #8: Line 289 the section title should not be the estimation of uptake coefficients of O₃ and N₂O₅, but the influence of the change of these parameters.

Response #8: The title is revised as ‘Impact of uptake coefficients of O₃ and N₂O₅ on chlorine species and nitrate’