Response to the Comments of Referee

Journal: Atmospheric Chemistry and Physics

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Title: Analysis of new particle formation events and comparisons to simulations of particle number concentrations based on GEOS-Chem/APM in Beijing, China

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We thank the reviewer and editor for providing helpful comments to improve the manuscript. We have revised the manuscript according to the comments and suggestions of the referee.

The referee's comments are reproduced (black) along with our replies (blue). All the authors have read the revised manuscript and agreed with the submission in its revised form.

< Anonymous Referee >

Comment: The manuscript has improved a lot and been corrected according to the comments. All the issues raised by the reviewers has been addressed point by point. I recommend this paper can be accepted after the minor revisions as given below.

Thanks for the suggestions and corrections, we have revised the manuscript accordingly.

Comment 1: The authors have fully discussed the limitation and uncertainties of the model result in this study. as you mentioned the H_2SO_4-DMA nucleation is probably the major path way of nucleation in Beijing (Yan et al., 2021; Cai et al.,2021,2022), and in your work, TIMN scheme simulation agree well with the nucleation. So how is the atmospheric implication of TIMN in Beijing?

In real atmosphere, there is probably more than one nucleation mechanisms occurring in some cases. Overall, TIMN agree better than other nucleation schemes used in this study, but one can easily see that TIMN simulation obviously underestimated the observed nucleation rate on March 13th (> 150 cm^{-3} s^{-1}), which may imply that other nucleation schemes which have not been included in the simulations, e.g., H_2SO_4-DMA or H_2SO_4-organics, may also simultaneously contribute to nucleation in this case.

And have evaluated the contribution by "ion-mediated process" in Beijing?

Sorry for misleading. We meant that four nucleation schemes including ion-mediated nucleation have been evaluated.

Comment 2: Line 17, to be favorable for

Corrected.

Comment 3: Line 55-56, please add the references to support the first sentence in this paragraph

Thanks for the suggestions. We have added references in the revised manuscript.

Comment 4: Line 60-63, can you give the suitable conditions for each nucleation mechanism, as you mentioned HIO_3 nucleation is dominant for coastal areas.

Thanks for the suggestions. H_2SO_4-H_2O binary theory usually predicts the nucleation rates at low temperatures, high relative humidities, small pre-existing aerosol concentrations and high sulfuric acid concentrations (Kulmala et al., 1998, 2000). For NH_3 mixing ratios exceeding about 1 ppt, the H_2SO_4-NH_3-H_2O ternary nucleation enhances the binary H_2SO_4-H_2O nucleation rate by several orders of magnitude (Korhonen et al., 1999). Ion-mediated nucleation mechanism provides a consistent explanation for a variety of tropospheric observations (Yu and Turco, 2000). Organics-mediated nucleation can explain NPF in some polluted areas (Wang et al., 2015).
These sentences have been added to the revised manuscript.

Comment 5: Line 188-189, I don't think the resolution of sulfuric acid can be 1 molecule/cm3, I recommend the digitals are given as $(6.1\pm3.1)*10^5$

Done as suggested.

Comment 6: Line 207, governed by aerosol Fuchs surface area, which is a representative parameter of coagulation scavenging (Cai et al., 2017a).

Corrected.

Comment 7: Line 216, necessary condition for NPF occurrence in the atmosphere

Corrected.

Comment 8: Line 329, can not

Corrected.

Comment 9: Line 420-425, does the APM model consider the nucleated particle growth process when simulate the PM mass concentration? If it is possible to evaluated the contribution of NPF to particle mass?

The APM model consider the contribution of nucleated particle growth process to total PM mass concentration.
In the atmosphere, the surface area of pre-existing particles not only serves as a coagulation sink but also as a condensation sink for precursor gases. The APM model takes into account the effect of surface area of pre-existing particles (Yu et al., 2020). In the model, the precursor gases that do not involve in the growth of nucleated particles (for cases with very small nucleation rates) condense on pre-existing particles instead so $PM_{2.5}$ and PM_{10} mass concentrations are close for all four schemes.
These sentences have been added to the revised manuscript.

Reference

Korhonen, P., Kulmala, M., Laaksonen, A., Viisanen, Y., McGraw, R., and Seinfeld, J.: Ternary nucleation of H_2SO_4, NH_3, and H_2O in the atmosphere, J. Geophys. Res., 104, 26349-26353, https://doi.org/10.1029/1999jd900784, 1999.

Kulmala, M., Laaksonen, A., and Pirjola, L.: Parameterizations for sulfuric acid/water nucleation rates, J. Geophys. Res., 103, 8301-8307, https://doi.org/10.1029/97jd03718, 1998.

Kulmala, M., Pirjola, L., and Mäkelä, J.: Stable sulphate clusters as a source of new atmospheric particles, *Nature*, 404, 66–69, https://doi.org/10.1038/35003550, 2000.

Wang, Z., Hu, M., Pei, X., Zhang, R., Paasonen, P., Zheng, J., Yue, D., Wu, Z., Boy, M., and Wiedensohler, A.: Connection of organics to atmospheric new particle formation and growth at an urban site of Beijing, Atmos. Environ., 103, 7-17, https://doi.org/10.1016/j.atmosenv.2014.11.069, 2015.

Yu, F. and Turco, R.: Ultrafine aerosol formation via ion-mediated nucleation, Geophys. Res. Lett., 27(6), 883-886, https://doi.org/10.1029/1999gl011151, 2000.

Yu, F., Nadykto, A. B., Luo, G., and Herb, J.: H_2SO_4–H_2O binary and H_2SO_4–H_2O–NH_3 ternary homogeneous and ion-mediated nucleation: lookup tables version 1.0 for 3-D modeling application, Geosci. Model Dev., 13, 2663–2670, https://doi.org/10.5194/gmd-13-2663-2020, 2020.