Response to the Comments of Referees

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Title: Analysis of new particle nucleation events and comparisons to simulations of particle number concentrations based on GEOS-Chem/APM in Beijing, China
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We thank the reviewers and editor for providing helpful comments to improve the manuscript. We have revised the manuscript according to the comments and suggestions of the referees.
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 #2 >

Comment: This study simulates NPF events in Beijing by applying GEOS-Chem/APM model, considering four nucleation mechanisms. It improved our understanding on nucleation and influence by meteorological factors. The TIMN nucleation scheme can predict nucleation well, however, more direct measurement data needed for validation the modeling results. The effect of meteorological conditions and precursors on nucleation should be further discussed in details. This paper is well organized and written. I recommend it can be accepted after the following major revisions.

We thank the referee for the positive comments on our manuscript. The manuscript has been carefully revised according to the referee's comments and suggestions.

Major issues
Comment 1: Line 113, The nucleation mechanism used in APM model is IMN parameterization scheme, which is developed based on the measurement and laboratory data elsewhere (Yu, 2006b). However, whether the parameterization is applicable in urban Beijing, with the high pollution level and unclear role that organics take place?

The reviewer raised a good point. Nucleation is a key process controlling particle properties in the atmosphere. To better understand the formation and evolution mechanisms of air pollution, especially in heavy pollution areas, it is necessary to assess the applicability of nucleation parametrizations currently available. This study aims to evaluate the performance of four currently widely-used nucleation schemes, and provide some clues on the contribution of different nucleation pathways to aerosol number concentrations.
The nucleation rates of different schemes in this study were calculated using lookup tables, which captured well the absolute values of nucleation rates and their dependence on key controlling parameters as observed during the well-controlled Cosmics Leaving

Outdoor Droplets (CLOUD) experiments (Yu et al., 2020). According to our simulations, the parameterization schemes can capture some new particle formation events in urban Beijing and provide a basis for discussing the new particle nucleation mechanism in urban areas. We acknowledge that it is possible other nucleation mechanisms such as H_2SO_4-amine and H_2SO_4-organics nucleations may also simultaneously contribute to nucleation in polluted urban areas, which needs further study in the future.

Can you talk about the uncertainties or bias of the simulation result due to the four parameterizations?

Some uncertainties may exist in nucleation schemes as a result of uncertainties in the thermodynamics data used in the nucleation model. There are six parameters controlling nucleation rates for TIMN (J_{TIMN}), including sulfuric acid vapor concentration ([H_2SO_4]), ammonia gas concentration ([NH_3]), temperature (T), relative humidity (RH), ionization rate (Q), and surface area of pre-existing particles (S). Compared to J_{TIMN}, there is one fewer controlling parameter for nucleation rates for THN (J_{THN}) (no Q dependence) and BIMN (J_{BIMN}) (no [NH_3] dependence), while nucleation rate for BHN (J_{BHN}) only depends on four parameters ([H_2SO_4], T, RH, and S). The uncertainties in the values of these parameters simulated by the model, as a result of uncertainties in the emissions, chemistry, and meteorology, will affect the simulation results. In addition, the real nucleation mechanisms in the atmosphere are complex and may involve additional parameters. Besides, the comparison between simulation results based on large grids and measurements also created uncertainties.
These sentences have been added to the revised manuscript.

Comment 2: Figure 2, can you explain why there no clear difference of sulfuric acid concentration between NPF and non-NPF days. Table 1 is not necessary as only two numbers are given. It can be given in the text, and better to give the mean ± standard deviation.

Thanks for the suggestions, we have deleted Table 1 and corrected the mean ± standard deviation.
According to Cai et al. (2017), sulfuric acid in Beijing during the campaign period was sufficiently high for nucleation events to occur and NPF events appeared to be governed by aerosol Fuchs surface area. The presence of gaseous sulfuric acid in concentrations exceeding 10^5 molecules cm^{-3} has been shown as a necessary condition to occur NPF in the atmosphere (Weber et al., 1999; Nieminen et al., 2009). Yan et al. (2021) found that observed sulfuric acid concentrations were higher on non-NPF days than on NPF days in the winter of 2018. Therefore, in this study, it is reasonable that sulfuric acid concentrations on NPF and non-NPF days were close.

In addition, as the authors mentioned, the sulfuric acid reported in this study is lower than the other studies, can you give the concentration level given by other studies? As

SO_2 decreased recent year in Beijing, the comparison should be conducted at the recent years, and also differed by seasons.

The daily maximum sulfuric acid concentrations measured in this study (> 10^6 cm^{-3}) are lower than those in summer of 2017 in Beijing (> 10^7 cm^{-3}) (Wu et al., 2020) and close to those in winter of 2019 in Beijing (> 10^6 cm^{-3}) (Foreback et al., 2022). This might be caused by the relatively weak solar radiation intensity in springtime and wintertime measurements compared with summertime observation.
We have revised the corresponding content of the manuscript.

Comment 3: Line 214, some studies reported that temperature can influence the NH_3 stabilizing with sulfuric acid, which finally affect the nucleation rate. However, in this work, it cannot be concluded the roles of temperature. In Beijing, NPF occurs more frequent in spring, winter than summer. The higher temperature on NPF days probably related with stronger solar radiation on clear days. It is difficult to evaluate the roles of temperature, as temperature, RH and solar radiation correlated under the similar synoptic conditions. As well as in line 228, I don't think a simple metrological factor, RH or solar radiation, can explain the NPF reasonably (such as, high RH usually occurs under cloudy days with low solar radiation). The meteorological factors have systematically influence on NPF.

Thanks for the comments. We agree that meteorological factors have systematical influences on NPF and it is difficult to isolate the effect of single factor on NPF.
We have revised the corresponding content of the manuscript and deleted Figures 4 and 5.

Comment 4: Figure 7, can you explain why modeled RH is much lower than the observed value? It this reasonable with the model uncertainties? For example, on March 26 and 27, the bias can be 10 K.

Temperatures (at the first layer, about 70 meters above the surface) input to the model are significantly lower than measurements taken near the surface. We compared temperatures (at 10 m above the displacement height) input to the model with measurements. As shown in Figure R1, the temporal variations of the temperature input to the model were more consistent with observations than before. Besides, the coarse spatial resolution may also cause the bias because of the "urban heat island" effect.

We have revised the Figure as Figure R1.

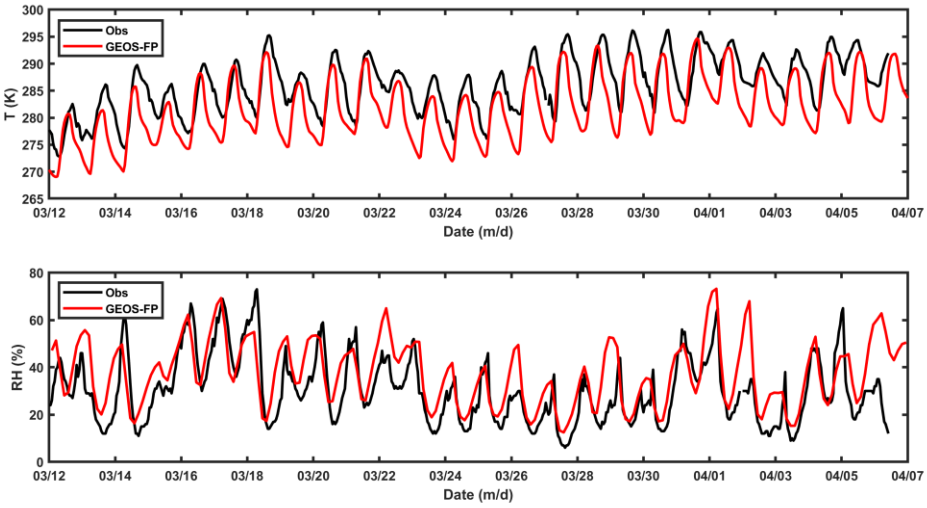

Figure R1. Time series of temperature (at 10 m above the displacement height) and RH (at the first layer, about 70 meters) input to the model and time series of measurements at a height of about 10 meters during the 26-day campaign.

Comment 5: Figure 10, can you calculate the observed nucleation rate, as compared with the simulated nucleation rate.

Thanks for your suggestion. We have added the measured particle nucleation rates reported in Cai et al. (2017) to Figure R2. The simulated nucleation rates in Figure R2 were lower than measured particle nucleation rates reported in Cai et al. (2017). The relatively coarse spatial resolution in a global model implies that the model produces a regional mean nucleation rate compared to the observation. Thus, it is difficult to perfectly reproduce the nucleation rate characteristics over urban areas. Moreover, we acknowledge that it is possible other nucleation mechanisms such as H_2SO_4-amine and H_2SO_4-organics nucleations may also simultaneously contribute to nucleation in polluted urban areas, which needs further study in the future.

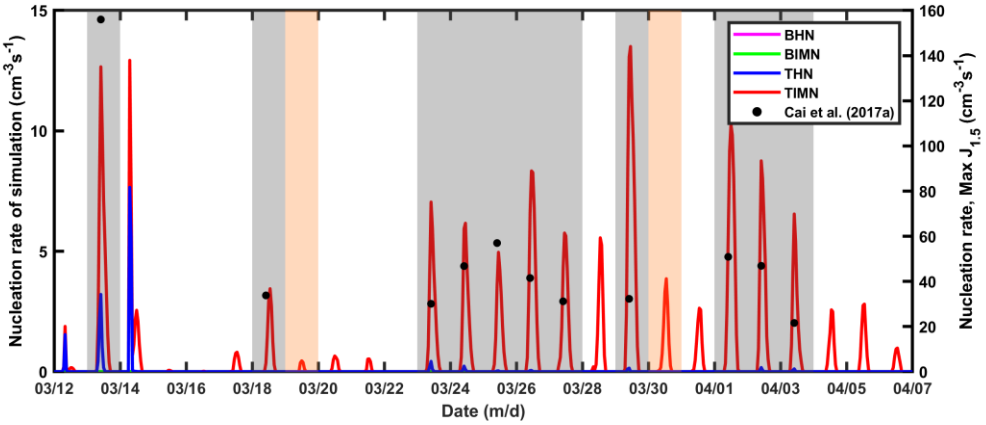

Figure R2. Time series of nucleation rates simulated on the basis of BHN, BIMN, THN, and TIMN schemes during the measurement period and the measured particle nucleation rates of 1.5 nm

particles (Max $J_{1.5}$) reported in Cai et al. (2017). The identified NPF days and undefined days are shadowed by grey and orange background, respectively.

Figure 11, the vertical distribution of nucleation rate has large uncertainties, and even no vertical data can validate the model result. I don't think it is robust confidence to represent the nucleation rates in the upper boundary layer in line 297.

We have deleted Figure 11 and the related discussion.

Comment 6: Can you model the sulfuric acid and validate the results by the measurement data? This can improve the confidence of model results.

Thanks for your suggestion. We believe that it may be more reasonable to compare the simulated sulfuric acid concentrations by higher resolution with the field observations. Because the simulated sulfuric acid concentration in a large grid cannot represent the measured sulfuric acid at a site in urban Beijing. We can discuss this problem in future work.

Minor problems:

Comment 7: Line 31-34, First, "new particle nucleation" is a repetitive phrase, normally we call this phenomenon "new particle formation", which includes nucleation and growth process. For the second sentence, the nucleated particles undergone condensation and coagulation processes and grow into larger sizes. However, water absorption is an independent process that characterize the particle hygroscopicity, which should not be included in the new particle formation process.

Thanks. Revised as suggested.

Comment 8: Line 99 and 101, for the data sources from website, the latest access time should be given.

Corrected.

Comment 9: Figure 1, the contour plot of PNSD near the detection limit below 5 nm looks wired, it seems only data of NPF was given. How is the data on other days? It is zero or has been excluded from the dataset? The author should provide the details about how to handle the PNSD data.

Sorry for the confusion. The plot near the detection limit below 3 nm in Figure 1 looks wired, possibly because the instrument for measuring sub-3 nm particles is different from the instrument for observing 3 nm to 10 μm (which is beyond the scope of this study).
We did not exclude the data on other days. Except for NPF days, the number concentrations of sub-3 nm particles on other days are very low and almost zero.

Comment 10: Line 213, 8 to 16:00 UTC or Local time, o'clock is not a formal written language.

Corrected.

Comment 11: Table 2, the restricted conditions (RH and solar radiation) proposed for identifying NPF only based on one-month measurement data is not robust. Even at the same location, the criterion can be changed due to seasonal variation of meteorological factors.

Thanks for the comment which we agree. We hope to conduct such analysis once more measurement data are available in the future, in order to improve our understanding. Table 2 has been deleted.

Comment 12: Figure 13, APM model cannot capture the peaks of PM_5 and PM_{10}, especially the severe pollution episode from March 14 to 18, is this due to the model spatial resolution or the emission inventory uncertainty?

Yes, we think that both the model spatial resolution and the emission inventory are potential factors to affect the results. We will discuss these issues in the future study. We have added these discussions to the revised manuscript.

Reference

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