We thank the reviewer for the comprehensive comments to help us improve the manuscript. Please see the detailed responses to your comments below.

The motivation behind this study is to update the nucleation and growth parameterizations in the WRF-Chem model, enabling it to simulate the particle formation and CCN formation processes in a coastal city in China. While some earlier studies have found a negative contribution of NPF to CCN, this study finds a positive contribution of NPF to CCN by adjusting the SI-SOA yield. The major updates include changes to key parameters, such as the H_2SO_4 accommodation rate, the HNO_3 condensation rate, the direct emission of primary organic aerosol, and, most importantly, the SI-SOA yield. This type of work is encouraged and fits within the scope of ACP.

[disclaimer: I'm not an expert who can judge whether the authors' model and setups represent the most advanced knowledge in their community.]

Major comments:

1. The authors should directly change the mass accommodation coefficient of H_2SO_4 from 0.1 to 1. There is enough experimental evidence showing this is the case. All other results should be revised with respect to this change. See 10.5194/acp-20-7359-2020

Thanks for the suggestions. In order to incorporate this suggestion and address the comments below, we have added another scheme (kinetics) in which the mass accommodation coefficient of H_2SO_4 is set to 1.0. While completely repeating all experiments would take tremendous of time and computational resources, we have added a number of simulations for the model evaluation and comparison. Therefore, the revised layout of the manuscript is to keep the original structure, but add the discussions based on kinetics scheme at the end of the manuscript.

2. L263: I believe it's time for everyone to stop using the activation scheme, given that the studies supporting it are from 2006-2007 and even the authors themselves may have moved on. Moreover, the dependence of J on H_2SO_4 is evidently non-linear. Several

studies in Chinese megacities have demonstrated the significance of H_2SO_4-DMA nucleation. While the situation might be different in a coastal city, it's unlikely that there is no NH_3 present. Incorporating DMA and NH_3 into WRF-chem may be challenging, as their sources may not be explicitly described. However, I encourage the authors to employ the H_2SO_4- NH_3 nucleation mechanism and rates in their study. They could use an estimated NH_3 concentration, as the nucleation rate from H2SO4-NH3 is less dependent on NH_3 than on H2SO4. The authors should compare the results obtained using the activation scheme with those obtained using the H2SO4-NH3 mechanism. If the latter yields superior results, it should be used as the default for other sensitivity tests. Conversely, if the H_2SO_4-NH_3 mechanism does not improve the results, this issue should be discussed. The problem may lie in other less certain modules instead of this experimentally confirmed mechanism.

Thanks for the suggestions and comments. As was described in the response to the first comment, we have added a number of new simulations. For instance, considering the reviewer's concern about the nonlinearity of the dependence of J on H_2SO_4, we added another nucleation scheme of kinetics, which assumes that the nucleation rate is proportional to the square of the concentration of sulfuric acid ($J = K[H_2SO_4]^2$). In this scheme, the mass accommodation coefficient of H_2SO_4 is set to one, and all the adjustment discussed in the linear-H_2SO_4 has been added in this scheme. The resulting simulated results are comparable to those obtained by the linear-H_2SO_4 nucleation mechanism. In addition, we conducted another set of simulations with H_2SO_4-NH_3 nucleation scheme, and the comparison indicates that the simulations under this scheme substantially overestimate the particle number concentrations. The reason likely lies in, that the reviewer has mentioned, the H_2SO_4-DMA was recently proposed to be the major nucleation scheme in megacities of China. To this end, we have added the relevant discussions in supplementary section S2. The section S2 is shown below as well.

To further verify the robustness of the model improvement in reproducing the observations, we select another empirical scheme, e.g., kinetics, nucleation for

evaluation. The repeated analysis for the smaller particle number concentrations (CN_{10-40}) indicates comparable performance between kinetics and activation schemes (Fig. S9), both showing improvement when mass accommodation coefficient is increased from 0.1 to 0.65. Considering that the mass accommodation coefficient is suggested to reach one in some studies (Stolzenburg et al., 2020), we therefore conduct another simulation under the kinetics nucleation scheme by increasing the mass accommodation coefficient to 1.0 (purple lines in Fig. 1), yielding comparable performance but with negative mean fractional bias contrasting to the positive one based on mass accommodation coefficient of 0.65 (green lines in Fig. 1; Table S1). For the large particle number concentrations (CN_{40-100}), the adjusted mass accommodation coefficient (1.0) together with low yield of SI-SOA at kinetics scheme shows similar improvements as activation (Fig. 2 and Table S2).
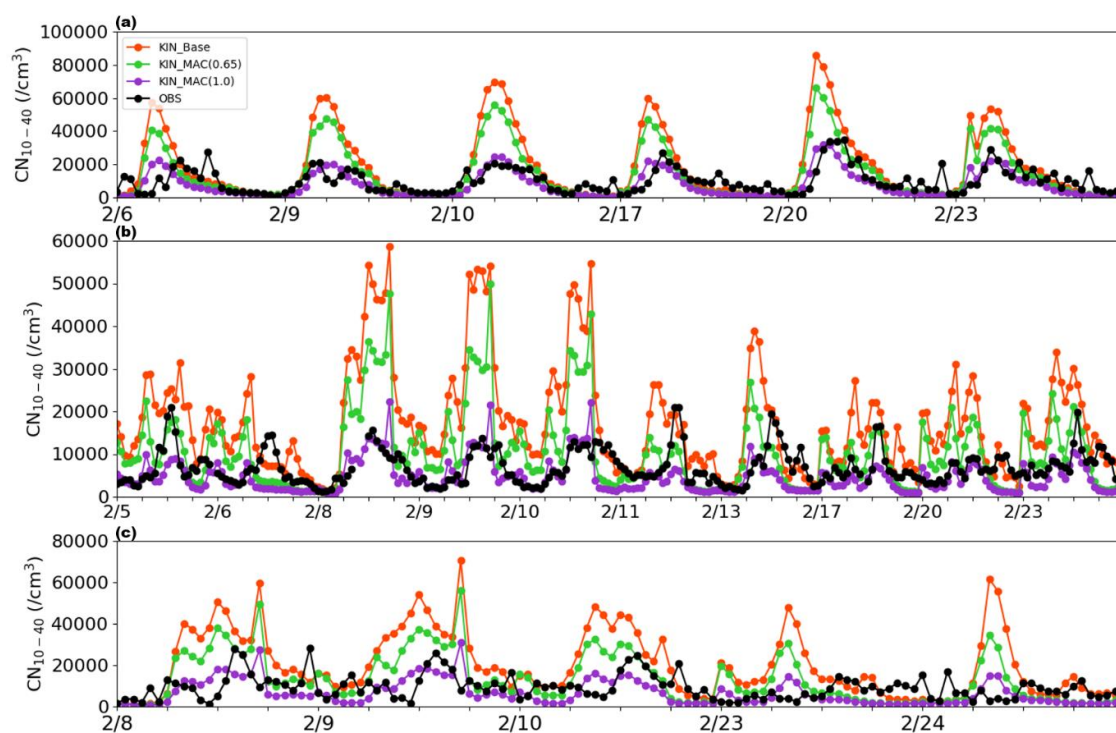

Fig. 1. The time series of CN_{10-40} on NPF days in (a) Qingdao, (b) Beijing and (c) Gucheng on February 5-24 simulated by Base (marked in orange) and MAC (green and purple lines corresponding to sulfuric acid mass coefficient of 0.65 and 1, respectively) using kinetics nucleation scheme (KIN) as well as from observations (OBS) (marked in black). All times are local times (LT).

Table 1 The statistics of model simulation and observation data for CN_{10-40} in Qingdao, Beijing and Gucheng

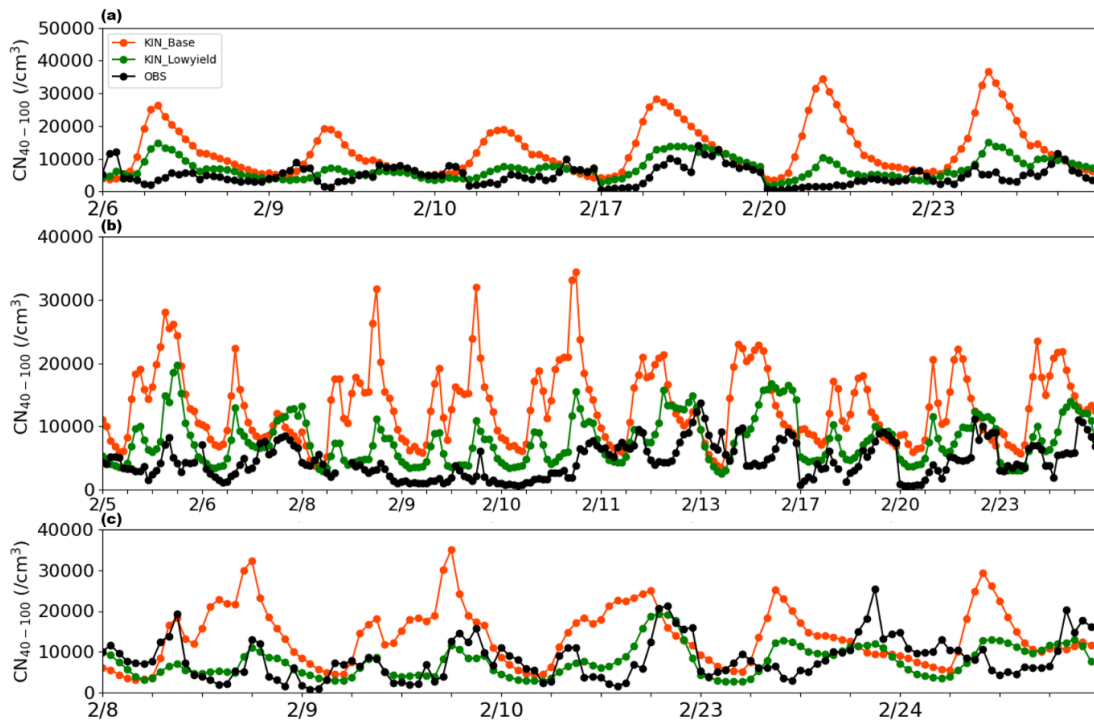| Observational sites / Simulation	Qingdao			Beijing			Gucheng		
	MFB (%)	MFE (%)	R	MFB (%)	MFE (%)	R	MFB (%)	MFE (%)	R
ACT_Base	48%	66%	0.69	81	90	0.35	62	82	0.21
ACT_ MAC(0.65)	1%	49%	0.70	23	65	0.39	11	67	0.13
KIN_Base	58%	83%	0.60	86	91	0.41	76	93	0.13
KIN_ MAC(0.65)	40%	71%	0.60	41	78	0.34	37	81	0.18
KIN_MAC(1.0)	-30%	57%	0.69	-40	61	0.41	-34	81	0.23

Fig. 2. The time series of CN_{40-100} on NPF days in (a) Qingdao, (b) Beijing and (c) Gucheng on February 5-24 simulated by Base (marked in orange) and Low_yield (marked in dark green) using kinetics nucleation scheme (KIN) as well as from observations (OBS) (marked in black). All times are local times (LT).

Table 2 The statistics of model simulation and observation data for CN_{40-100} in Qingdao, Beijing and Gucheng.

Observational sites / Simulation	Qingdao			Beijing			Gucheng		
	MFB (%)	MFE (%)	R	MFB (%)	MFE (%)	R	MFB (%)	MFE (%)	R
ACT_Base	98	102	0	103	106	0	50	72	0
ACT_Lowyield	32	53	0.42	59	65	0.47	-5	47	0.46
KIN_Base	88	94	0	97	100	0	50	74	0
KIN_Lowyield	36	52	0.39	53	60	0.49	-7	48	0.46

Following the empirical nucleation scheme, we then conduct a classical nucleation mechanism to take both chemical species and meteorological conditions directly into account (Sihto et al., 2006). For instance, we select a commonly used H_2SO_4-H_2O-NH_3 ternary homogeneous nucleation which is highly dependent on temperature and relative humidity (Napari et al., 2002). The number concentrations at 10–40 nm are much higher (Fig. 3), at either low or high mass accommodation coefficient, compared to observations and the empirical schemes abovementioned, and the diminished effect during the adjustment of mass accommodation coefficient is likely a result of NH_3.
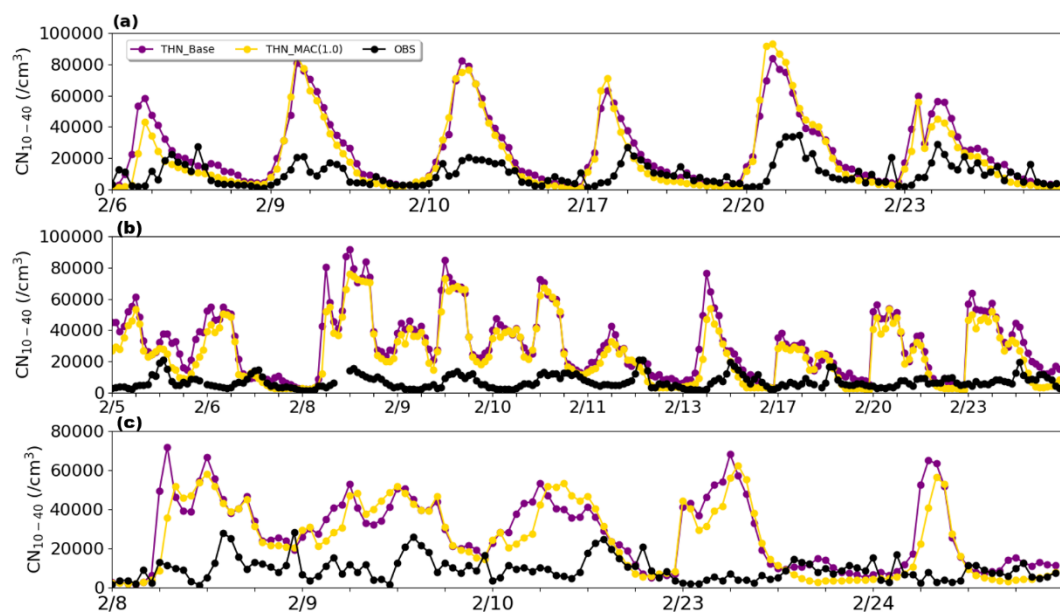
Fig. 3. The time series of CN_{10-40} on NPF days in (a) Qingdao, (b) Beijing and (c) Gucheng on February 5-24 simulated by Base (marked in purple) and MAC (marked in yellow) using H_2SO_4-H_2O-NH_3 ternary homogeneous nucleation (THN) as well as from observations (OBS) (marked in black). All times are local times (LT).

Contrasting to the scheme of H_2SO_4-H_2O-NH_3, the formation of sulfuric acid (SA)-dimethylamine (DMA)-water clusters has been found to be important sources of new particle formation in megacities over China (Yao et al., 2018). Bergman et al. (2015) applied amine-enhanced nucleation parameterization to an aerosol climate model to estimate the effect of amine on new particle formation on a global scale, indicating that high nucleation rates are confined to regions close to the amine source due to the short lifetime of amines. Because of the short life of amines, the emission of amines remains to be highly uncertain and deserves further investigation (Chang et al., 2021). By comparing this classical nucleation scheme with the empirical one (e.g., kinetics), the spatial distibutions of particle formation rate between these two types of nucleation schemes are largely consistent.

3. I find Session 3.4 to be particularly fascinating, but it's currently buried amidst a lot of less significant information. This session should be considered one of the key findings of this study and given prominence in both the abstract and conclusion. The

yield of SI-SOA remains highly uncertain, and I'm surprised to learn that such a small change in the reaction rate coefficient can have such a significant impact on the contribution of NPF to CCN. I hadn't expected this result at all. This finding underscores the need for further research into SI-SOA yield in polluted environments, particularly since urban environments are highly complex and model treatments are often oversimplified. Clearly, a better understanding of NPF's contribution to CCN hinges on a better grasp of this prerequisite knowledge.

We thank the reviewer for the positive comment on the discussion of session 3.4. We have revised the discussion based on the reviewer's suggestion. As the reviewer pointed out the uncertainty, actually when we apply the quadratic-H_2SO_4 nucleation scheme, the sensitivity numerical simulations with high and low yield of SI-SOA result in comparable contributions to CCN, which differs from the results using the linear-H_2SO_4 nucleation scheme. It may be related to particulate hygroscopicity dependence on the nucleation scheme. More future studies are necessary to investigate this issue. As was suggested by the reviewer, more work is needed to improve understanding of NPF's contribution to CCN.

Minor comments:

L39-41: a number of observations may be misleading. There are more than enough observations showing the positive correlation of NPF and CCN. Additionally, while some simulations do not show positive correlation of NPF and CCN in a global scale, many of the models do. If the authors' statement is about e.g., polluted environments or more specific the Chinese city, the authors should clearly be stating so. Otherwise they should modify this sentence properly.

Thanks for the suggestion. We have rephrased the descriptions. The words of "polluted environment" has been added to constrain the descriptions of negative contribution of NPF to CCN.

L74: high-efficiency nucleation efficient nucleation

Done.

L110-121: it appears the authors are only talking about WRF-chem. They should carefully mention this clearly in the manuscript that it is the WRF-chem model, not a general "model" that is observing negative correlation between NPF and CCN.

Done. We have refined the description of the model in the manuscript.

L190: change C* to $c*$ (italic, lower case) throughout the manuscript.

Done.

L199: Please write explicitly the used equation and all the parameters.

Done.

L333: try to reduce using MFB etc. Use the abbreviations only for models runs otherwise readers easily get confused.

We have reduced the abbreviations in the revised manuscript.

L347: space between $\mu g\ m^{-3}$play

Revised.

L349: Does this mean that all the vapours only have two different volatilities?

The volatilities mentioned in this sentence mainly indicate that for primary organic aerosol, with effective saturation concentration ($c*$) of POA is 10^{-2} and 10^{5} $\mu g\ m^{-3}$, respectively. For gas phase SOA oxidized by volatile organic compounds, the effective saturation concentration is set to be one, which is not the focus of the study, therefore, we did not mention the volatility of this part SOA. To make it clear, we have deleted the descriptions of two sets of volatilities, and directly discuss the source of primary organic aerosol with effective saturation concentration at 10^{-2} $\mu g\ m^{-3}$.

L354: Gas phase POA forms close to the emission source. However, with…

 Done.

L356: Therefore, POA may not contribute to particle growth away from the emission sources. Or something similar.

 The sentence has been elaborated.

L364: Please label the Figure S1 panels. There are also clear signs of grey bars in the figure. Please remove those when putting the figures together.

 Done.

L412: Avoid using too many abbreviations (PNC).

 The abbreviations have been removed.

L416: Which nucleation mechanism is used in this study?

 Organics nucleation mechanism is used in this study, which has been added in the revised manuscript.

L506: Are there measurement data for figures d,e,f?

 We have observations for particle number concentrations over these three bins. Since we have done particle number concentration evaluations in Fig. 3, we try not to repeat the information in this figure.

L538: This session is very interesting. I think this is worthy to be emphasised.

 Thanks for your suggestion. We have elaborated the writing, and added another scheme (kinetics) to further support the finding.

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