

### **Response to Reviewer #3 (our response in colour)**

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

#### **Reviewer #3:**

This study improves the simulation accuracy of particle and CCN number concentration through adjusting serval parameters such as the mass accommodation coefficient. The results are interesting and has a good potential to improve the model of NPF and its impact on CCN number concentration. This study is within the scope of the journal ACP. I recommend this paper for publication after the following issues are resolved.

#### **Major comments:**

1. Line 147. Li et al (2015) shows the measurement of bulk CCN, not the measurement of size-resolved CCN. The authors should give more information about the size-resolved CCN measurement, including the flow set, multi-charge calibration method, data quality control and so on.

Thank you for your comments. We have revised the observation method of CCN in section 2.1 of the manuscript. The observations of bulk CCN refer to the study of Li et al. (2015).

2. Line 262: The extremely low volatility volatile organic compounds (ELVOCs) can also have a contribution for the nucleation events.

Right, extremely low volatile organic vapors can be involved in particle nucleation. In this study, the empirical nucleation scheme is strongly governed by the nucleation coefficient, which indeed is a combination of multiple chemical species with different volatilities, which should include the extremely low volatility volatile organic compounds.

2. Line 458-460: The activation critical particle size at SS=0.2% is about 120~130 nm. It is interesting that the  $CCN_{0.2\%}$  has an obvious underestimation when  $CN_{10-1000nm}$  has an overestimation (Figure 3c) for the RACD scheme. The change of activation ratio ( $N_{CCN}/N_{CN}$ ) can be further analyzed.

Thanks for your comments and we add the analysis of the activation ratio. The results are consistent with the trend of CCN, in general showing similar features as CCN.

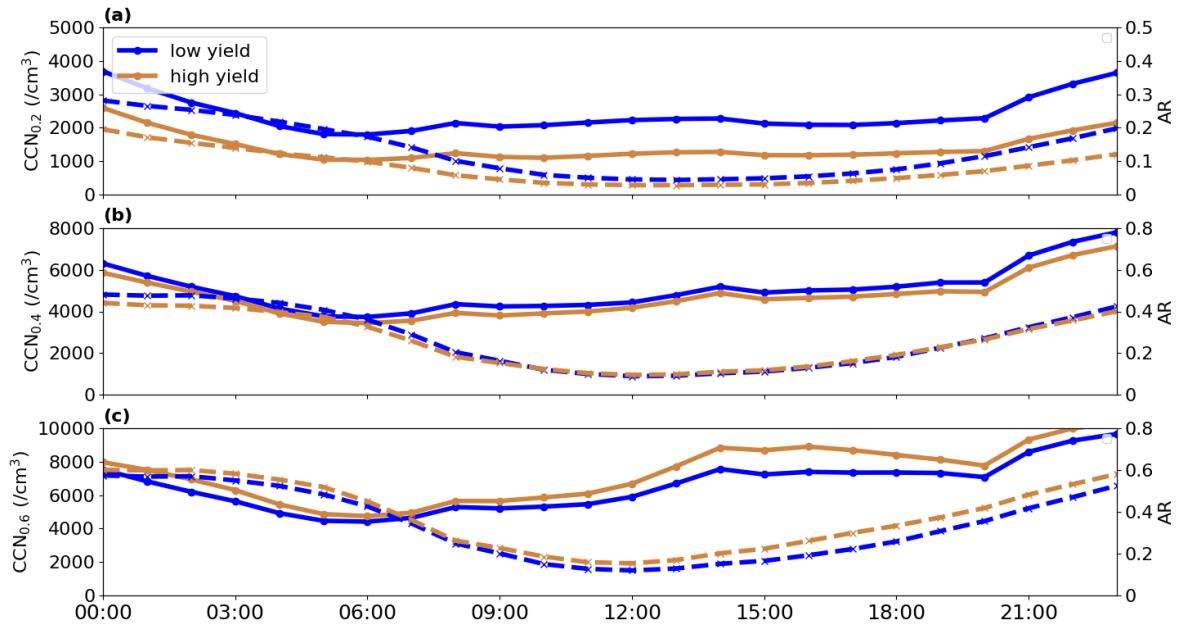


Fig. 1. Average diurnal variation of (a)  $\text{CCN}_{0.2\%}$ , (b)  $\text{CCN}_{0.4\%}$  and (c)  $\text{CCN}_{0.6\%}$  on NPF days in Qingdao on February 5-24, 2017, in Low-yield and High-yield simulations, shown as blue and brown lines. The solid line representing CCN concentration and the dashed line corresponding activation ratio (AR).

The underestimation of CCN at low supersaturation (0.2%) concomitant with the overestimation of particles at 100–1000 nm under high yield SI-SOA, as mentioned by the reviewer, is believed to be influenced by the hygroscopicity of the particles in this study. Compared to the high yield of SI-SOA, the reduced yield of SI-SOA tends to decrease organic matter which enhances the hygroscopicity of the particles to be easier to activate to CCN, therefore, the bias of CCN simulation is substantially reduced. To verify this hypothesis, we calculated the hygroscopicity parameter ( $\kappa$ ) for 100–1000 nm particles, and the values increase from 0.26 at high SI-SOA yield to 0.30 at low SI-SOA yield, indicating the enhanced hygroscopicity at low SI-SOA yield.

## 1. Line471-473: Why secondary inorganic aerosols (SIA) is not the major contributor of condensational growth?

Different chemical species are involved in the growth of particles. Secondary inorganic aerosols (ammonium, nitrate, sulfate) and secondary organic aerosols all make important contributions to the growth of newly formed particles in the atmosphere (Xiao et al., 2015; Ehn et al., 2014; Zhu et al., 2019). However, their importance differs

dependent on the particle sizes. For particles less than 40 nm, the condensation of semi-volatile substances such as nitric acid on particles tends to be inhibited by the high acidity of small particles (Roldin et al., 2011; Deming and Ziemann, 2021). For instance, based on observed evidence, Li et al. (2022) found in Beijing in 2018 showed that organic matter is the main component of 8–40 nm particles, with mass fraction of  $80\pm8\%$ , followed by sulfates accounting for  $13\pm7\%$ , and inorganic nitrates accounting for less than 3%. Therefore, we emphasize the secondary inorganics may not play a major role for smaller particles.

2. Figure 5. The authors emphasize that the model modification has a largest impact on the simulation of CCN over the North China Plain. It is necessary to verify the result based on the measurement in this region.

Thanks for the suggestions. We have tried to achieve more data, and it turned out we got more data on particle number concentrations, but not CCN. Therefore, we added more evaluations of particle number concentrations over two more sites at North China Plain. The model evaluations (Section S2 in the supporting information) are in general consistent with that in Qingdao, warranting the confidence of the model used in the study.

**Technical suggestions:**

Line 249: “particles growth” should be “particle growth”

Done.

Line 269: “NFP” should be “NPF”

Revised.

Line 316-328: This paragraph use “ $\alpha$ ” and “MAC” as the abbreviation of “mass accommodation coefficient”. I suggest using a unified symbol in text

Done. We have reduced the use of abbreviations in the manuscript.

## References:

Deming B, Ziemann P 2021. Measurements of the partitioning of nitric acid and sulfuric acid in aqueous/organic phase-separated systems. *Environmental Science: Atmospheres* [J], 1(

Ehn M, Thornton J, Kleist E, et al. 2014. A large source of low-volatility secondary organic aerosol. *Nature* [J], 506(476-479).

Li K, Zhu Y, Gao H, et al. 2015. A comparative study of cloud condensation nuclei measured between non-heating and heating periods at a suburb site of Qingdao in the North China. *Atmospheric Environment* [J], 112(40-53).

Li X, Li Y, Cai R, et al. 2022. Insufficient Condensable Organic Vapors Lead to Slow Growth of New Particles in an Urban Environment. *Environmental Science & Technology* [J], 56(14): 9936-9946.

Roldin P, Swietlicki E, Schurgers G, et al. 2011. Development and evaluation of the aerosol dynamics and gas phase chemistry model ADCHEM. *Atmos. Chem. Phys.* [J], 11(12): 5867-5896.

Xiao S, Wang M Y, Yao L, et al. 2015. Strong atmospheric new particle formation in winter in urban Shanghai, China. *Atmos. Chem. Phys.* [J], 15(4): 1769-1781.

Zhu Y, Li K, Shen Y, et al. 2019. New particle formation in the marine atmosphere during seven cruise campaigns. *Atmos. Chem. Phys.* [J], 19(1): 89-113.