Response to the reviewers

Referee #2

The authors present a framework combining multi-source data with Random Forest Regression to predict CCN concentrations. The significant improvement in prediction accuracy over the traditional WRF-Chem model, particularly in capturing long-term trends in the North China Plain. This study bridges the observation gap and highlights the climate benefits of emission controls. The methodology is sound, and the results are compelling. I recommend publication after minor revisions.

Re: We deeply appreciate the time and effort that the reviewers have dedicated to evaluating our manuscript. We believe that the revised version has been greatly improved in terms of the rationality of the model development methodology and the reliability of the results.

1. Organic matter was found to be the most crucial indicator for the CCN concentration prediction. The results are interesting; the inorganic salts are always thought to have high potential for the CCN. More explanation is needed here, if the number concentrations would increase under the high OM conditions?

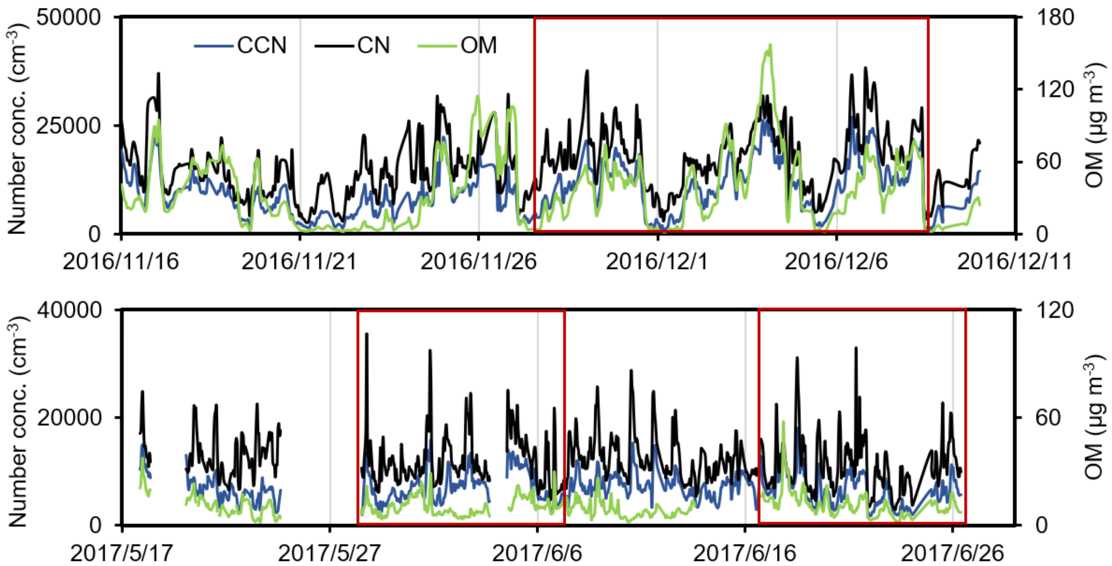

Fig. R1 Time series of CN, CCN number concentration and organic mass concentration in the campaigns of 2016BJ and 2017BJ.

Re: Thank you for your insightful comment. In this study, organic matter constitutes 20 – 90% of the aerosol mass in the North China Plain. Although OM is generally less hygroscopic than inorganic salts, its large mass fraction can compensate for its lower hygroscopicity, leading to a significant overall impact on CCN activation. In addition, recent studies have shown that OM can be partially hygroscopic or even surface-active, especially after aging and oxidation. Oxygenated organic aerosols can exhibit κ values comparable to those of some inorganic species. Moreover, OM can internally mix with inorganic salts, thereby altering the overall hygroscopicity of the particles. And we also noted that the CN number concentration would increase under the high OM conditions as shown in Fig. R1, further suggesting the OM can serve as the potential seed for CCN.

We have added some explanation as follows or see **Lines 236-254**:

"…Organic matter emerges as the most crucial indicator with the highest SHAP value. The wide range of SHAP values for OM reflects the diversity of its physicochemical properties. Specifically, low-concentration or freshly emitted hydrophobic OM contributes negatively to N_{CCN} (suppressing activation), whereas high-concentration or aged/oxidized OM contributes positively (promoting activation). From an overall perspective, SHAP values increase monotonically with OM concentration, and the absolute values of positive SHAP values exceed those of negative ones, demonstrating a synergistic positive effect of OM concentration on the variation of CCN number concentration. This finding differs from the conventional view that inorganic salts contribute more to CCN due to their strong hygroscopicity (Petters and Kreidenweis, 2007). However, in fact, we also note that under conditions of high OM, the concentrations of CN and CCN indeed show an increasing trend (Fig. S5). In addition, previous studies have shown that in the North China region where the proportion and concentration of OM are both high, organic particles affected by strong anthropogenic emission sources was found exhibit strong hygroscopicity, enabling them to serve as more effective CCN (Liu et al., 2021); in addition, the surface tension lowering effect of OM particles in this region can also enhance particle CCN activity (Fan et al., 2024). Therefore, the SHAP analysis results further confirm the conclusions of previous studies…"

2. The influence of temperature showed the bidirectional influence, whether the temperature would influence the emission sources or the chemical reaction in the atmosphere.

Re: Thanks for the comments. The SHAP analysis reveals a bidirectional nonlinear influence of temperature on CCN prediction, reflecting the dual regulatory role of temperature on emission sources and atmospheric chemical reactions. On the one hand, elevated temperatures promote VOC emissions and photochemical reactions, enhance the formation and aging of secondary organic aerosol (SOA), and increase particle hygroscopicity. On the other hand, low temperatures facilitate gas-particle partitioning, while high temperatures may accelerate new particle formation but suppress nucleation rates. These competing mechanisms collectively contribute to the observed bidirectional influence of temperature, which is consistent with the findings of Song et al. (2022) regarding the nonlinear modulation of CCN activity by temperature. We have added some explanation as follows or see **Lines 267-275**:

"…Temperature demonstrated a bidirectional influence, suggesting nonlinear modulation of CCN activity potentially associated with the temperature dependence of nucleation growth and secondary generation of particles (Song et al., 2022). Specifically, temperature-driven enhancements in emissions exacerbate the formation of secondary organic aerosols, thereby affecting CCN concentrations (Lian et al., 2025). In addition, temperature significantly influences aerosol formation, aging, and transformation processes by regulating photochemical reaction rates, particle aging processes, and gas-particle partitioning, which in turn affect nucleation processes and exert important impacts on CCN concentrations…"

3. The ratio between POA and SOA is important for the number of CCN. Can the model separate these two components? How about the performance of the simulation?

Re: Thank you for your suggestion. The RFRM model used in this study takes OM mass concentration as input and does not distinguish between primary organic aerosol (POA) and secondary organic aerosol (SOA). This is because we used the TAP dataset for spatiotemporal prediction, which does not provide detailed POA and SOA information. The high SHAP importance of OM indirectly suggests that incorporating POA and SOA information could have a significant impact on CCN prediction. In the future, introducing the POA/SOA ratio as a feature may further improve the model's interpretability.

4. A few sites near the coast with positive values in recent years, why accumulation-mode particles increase, more discussion would be helpful.

Re: The positive trends in accumulation-mode particles at coastal sites can be explained as: first, enhanced primary combustion emissions from sources other than on-road vehicles (e.g., industrial and residential coal burning) (Zhu et al., 2021); sea salt contributions a substantial source of accumulation-mode particles under marine air influence (Zou et al., 2024). And previous studies have demonstrated that baseline sea spray fluxes contribute approximately 50% of the submicron particle number flux in the accumulation mode even under clean marine conditions (Geever et al., 2005). Some explanations have been added as follows or see **Lines 400-408**:

"…Interestingly, note a few sites with positive values (upward trends in N_{CCN}) are mainly located along the coast. An increase in the fraction of accumulation-mode particles in coastal areas has been reported contributing more CCN (Zhu et al., 2021). In fact, previous studies have revealed that in Qingdao, enhanced primary combustion emissions from sources other than on-road vehicles (e.g., industrial and residential coal burning) serve as the main driver of increased accumulation-mode number concentrations (Zhu et al., 2021). Furthermore, coastal observations have identified sea salt aerosols as an important source of accumulation-mode particles (Zou et al., 2024) …"

References:

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