We are very grateful to the evaluations from the reviewers, which have allowed us to clarify and improve the manuscript. Below we addressed the reviewer comments, with the reviewer comments in black and our response in **blue**.

Reply for the referee comment#1
General Comments:

“The effect of organic nucleation on the indirect radiative forcing with a semi-explicit chemical mechanism for highly oxygenated organic molecules (HOMs)” by Shao et al. is a science development that evaluates the aerosol, cloud, and radiative forcing changes in the CAM-Chem model using a newly developed organic chemistry mechanism from Xu et al. (2022) and a new HOM nucleation scheme developed by Shao et al. (2024). The paper is succinct and has an overall organized structure, although I think some key information on the critical insights should be discussed earlier in the introduction section instead of in Sect. 4 (see the second comment below), especially because this insight appears throughout the whole manuscript but is not explained early on. Many comments of mine are requests for clarification. I would suggest major revision for this paper, which can be accepted after addressing the comments below. The manuscript can also be benefited by some rewriting to improve grammar and readability.

Response: We would like to thank the referee for providing the insightful suggestions, which indeed help us further improve the manuscript. We have added required discussion to account for the major and minor comments and marked the corresponding line number in the revised paper. Please see the revision and the response for the comments as follows.

Major Comment#1: Lines 28-29: The readability of the sentence is not good since the reader is not clear what Gordon 2016 concluded. It may make the sentence clearer by rephrasing the sentence as “The reduction is mainly driven by …, instead of the findings of Gordon et al. (2016) that the ~1 nm nucleation drives the reduction.”

Response: We apologize for any confusion caused by the original sentence. The sentence in Lines 28-29 were modified as (The underlined content is newly added or modified):

“~~Unlike the findings of Gordon et al. (2016), the reduction is mainly driven by a greater enhancement of the sub-20 nm growth rate (GR) in the PI atmosphere compared to PD instead of the ~1 nm nucleation rate ($j_{1.7\mathrm{nm}}$) .~~ The reduction is mainly driven by a greater enhancement of the sub-20 nm growth rate (GR) in the PI atmosphere compared to PD, instead of the findings of Gordon et al. (2016) that the ~1 nm nucleation rate ($j_{1.7\mathrm{nm}}$) drives the reduction.”

Major Comment#2: Line 48: new particle formation can be replaced by NPF.
Response: Done. Thanks.

Major Comment#3: Lines 72-73: It is not clear to the reader why there is a reduction in the magnitude of the radiative forcing. This is explained at the very end of the manuscript in Sect 4, but readers are confused here, so the importance of HOM-driven NPF in line 68 could be illustrated here in line 72. Some of the information in lines 280-285 could be covered here.

Response: To improve clarity, we have revised lines 72-73 to better explain the reduction in radiative forcing. This revision incorporates the importance of HOM-driven NPF and integrates key points from lines 280-285.

The sentence in Lines 68-73 were modified as (The underlined content is newly added or modified):

“HOM-driven NPF is especially important in the pristine PI atmosphere, where concentrations of sulfuric acid and ammonia were much lower. Simulations of the pre-industrial atmosphere form the baseline for calculating anthropogenic radiative forcing in global models (Carslaw et al., 2013), where higher monoterpene emissions led to greater HOM concentrations, thereby enhancing nucleation and particle growth. Using global model simulations, Gordon et al. (2016) showed that new particles formed from monoterpene-derived HOMs could increase CCN concentrations in the PI environment by 20% to 100%, a rise considerably larger than the increase simulated for PD conditions. This leads to a 27% reduction in negative radiative forcing ~~due to changes in cloud albedo~~ since 1750, decreasing by between -0.28 W m^{-2} and -0.06 W m^{-2}.”

Major Comment#4: Lines 81-83: The sentence is not clear here by themselves. Reader is curious and wants more details on what the specific advances in Xu 2022 and Shao 2024 are, and why they are better than Vehkamaki 2002, Gordon 2016, and other relevant paper. Please use a few sentences here to give a summary. You have more details in Sect. 2, but I think more details are needed here as well.

Response: Sorry for confusion caused by the original sentence. We have revised Lines 81-83 to provide more detailed information on the advances in Xu et al. (2022) and Shao et al. (2024). The sentence in Lines 81-83 were modified as (The underlined content is newly added or modified):

"Considering the unequivocal evidence for the role of biogenic organics in producing atmospheric particles, Shao et al. (2024) have recently incorporated a state-of-the-art representation of HOMs from various chamber experiments (Xu et al., 2022). This representation semi-explicitly treats the unimolecular autoxidation of monoterpene-derived RO_2 radicals and their self- and cross-reactions with other RO_2 species, rather than using the empirical fixed HOM yield (Gordon et al., 2016; Zhao et al., 2018). In addition, Shao et al. (2024) introduced a HOM-involving nucleation parameterization (Riccobono et al., 2014; Kirkby et al., 2006) and enabled these organics to condense onto newly formed sub-20 nm particles. ~~and update the nucleation scheme involving HOMs, ACCs, H_2SO_4, NH_3, and ions from previously published CLOUD chamber experiments in a global chemistry-climate model (Shao et al., 2024). Additionally, Shao et al. (2024) account for organics condensing on newly formed sub-20 nm particles~~."

Major Comment#5: Lines 177-178: Why is H_2SO_4 overestimated over Brazil, Barrow, and Graciosa but not other places? Please also explain why H_2SO_4 concentration is overestimated in CAMChem.

Response: Although direct observations of H_2SO_4 are not globally available, existing evaluations indicate that CAM-Chem systematically overestimates its concentration across many regions (He et al., 2014, Shao et al., 2024), not only in Brazil, Barrow, and Graciosa. This is a feature of CAM6, as evidenced by comparisons with previous model simulations (Table S6 in Shao et al. 2024) and measurements (Table S7 in Shao et al. 2024), which will be presented below.

We highlight that this overestimation has the largest impact in Brazil, Barrow, and Graciosa. At these urban sites the growth of newly formed particles is affected mainly by the condensation of H_2SO_4 onto sub-20 nm particles (see Shao et al., 2024, Fig. 7), so the overestimation of H_2SO_4 concentration directly amplifies the simulated growth rate. In rural and mountainous regions abundant monoterpenes supply organic vapours that dominate particle growth, so the H_2SO_4 bias has only a minor effect there.

Table S5 (from Shao et al., 2024). Comparison of Gas Burden (Tg S) across different studies

	This study (CESM-MAM4)	Liu et al. (2012)		Spracklen et al. (2005)	Mann et al. (2010)
	Inorg_Org	CESM-MAM3	CESM-MAM7	GLOMAP-bin	GLOMAP-mode
SO_2	0.29	0.35	0.34	0.49	0.3 (0.2-0.68)
DMS	0.067	0.067	0.067	0.04	0.027 (0.02-0.15)
H_2SO_4	0.00054	0.0004	0.00042	/	0.0001

Table S6 (from Shao et al., 2024). Comparison of H_2SO_4 concentration with measurements

Measurement site	$[H_2SO_4]$ 10^6 mole cm^{-3}			
	Mean		Median	
	Measurement	Simulation	Measurement	Simulation
Hyytiälä, Finland	0.43	2.61	0.18	0.3
San Pietro Capofiume, Italy	5.40	4.57	2.40	2.79
Melpitz, Germany	6.43	10.53	2.94	2.8
Niwot Ridge, Colorado USA	1.83	2.52	1.40	1.36
Atlanta, Georgia USA	12.90	9.95	2.85	1.03
Beijing, China	2.51	17.91	1.81	3.15

In CAM-Chem the overestimation of gas-phase H_2SO_4 originates from several reasons. Anthropogenic emissions of the SO_2 are likely too high, wet scavenging removes SO_2 too inefficiently, and the representation of in-cloud aqueous chemistry is overly simplified. The overestimation of SO_2 emission in CAM-Chem produced a normalized mean bias of 219.1 % for SO_2 concentration, rising to 244.7 % at CASTNET sites (He et al., 2015). Sensitivity experiments by He and Zhang (2014) show that reducing SO_2 emissions by 30 % lowers the simulated bias from 291.8 % to 152.2 %. Ge et al. (2021, 2022) further demonstrated that incorporating detailed in-cloud aqueous-phase chemistry and enhanced sulfate wet deposition significantly improves the simulation of SO_2 and H_2SO_4. In addition, He et al. (2015) reported that CAM-Chem tends to underestimate precipitation in the eastern United States, which weakens wet scavenging of SO_2 and amplifies the overestimation of H_2SO_4.

The sentence in Lines 177-178 were modified as (The underlined content is newly added or modified):

"Apart from urban regions like Manacapuru, Brazil, the overestimation also increases over oceanic regions such as Barrow and Graciosa. These overestimations in CCN numbers in the Inorg model are likely related to the overestimation of H_2SO_4 concentration in CAM6-Chem (Shao et al., 2024), as these regions are more sensitive to H_2SO_4 due to the limited presence of organic NPF precursors like monoterpenes. The overestimation of H_2SO_4 concentrations in the CAM-Chem model is likely the result of multiple contributing factors, such as overestimated SO_2 emissions (He et al., 2014; He and Zhang, 2014), insufficient representation of in-cloud chemistry (Ge et al., 2021; 2022), underestimated wet deposition processes (He et al., 2015; He and Zhang, 2014)."

Major Comment#6: Line 179-180: It is not clear whether the Shao 2024 organic nucleation scheme (Inorg_Org) has increased CCN across the whole globe compared to the Inorg run. It looks like some places have worsened underestimations (such as Azores for 0.5%SS and 1%SS), which requires an explanation.

Response: The organic NPF scheme in Shao et al. (2024) (Inorg_Org) does increase the global CCN burden compared to the Inorg simulation, but not necessarily the surface number concentration. The CCN values shown for the Azores in Figure 1 represent surface concentrations (Table 1); therefore, it is possible that the underestimation becomes more pronounced in the Inorg_Org simulation after including the organic NPF scheme.

The main reason for the worsened underestimations in Azores (ocean region) at 0.5% SS and 1% SS may be that, after including organic NPF, heteromolecular nucleation involving H_2SO_4 and organics ($J_{SA\text{-}Org}$) consumes more H_2SO_4 over land, as monoterpene emissions and the formation of HOMs are mainly concentrated over there (Fig. S3). As a result, less H_2SO_4 is transported to the ocean, where nucleation (Figs. S4 and S5) and sub-20 nm particle growth are primarily driven by H_2SO_4 (Fig. 7 in Shao et al., 2024), leading to decreased nucleation and growth rates over ocean.

In light of this, the sentence in Line 219 were modified as (The underlined content is newly added):

"The enhancement in nucleation rates due to the inclusion of organic nucleation is more significant in the PD experiment (39%) compared to the PI experiment (6%) (Fig. 4). This is mainly caused by higher sulfuric acid concentrations in the PD environment (Fig. S2), resulting in higher heteromolecular nucleation rates involving sulfuric acid and organics ($J_{SA\text{-}Org}$) (Figs. S6 and S7) over land, where both H_2SO_4 and HOMs show high values. Consequently, more H_2SO_4 is consumed over land (Fig. S16), reducing its transport to oceanic regions (Fig. S17). As a result, nucleation rates decrease over the ocean in both PD and PI experiments (Fig. 4)."

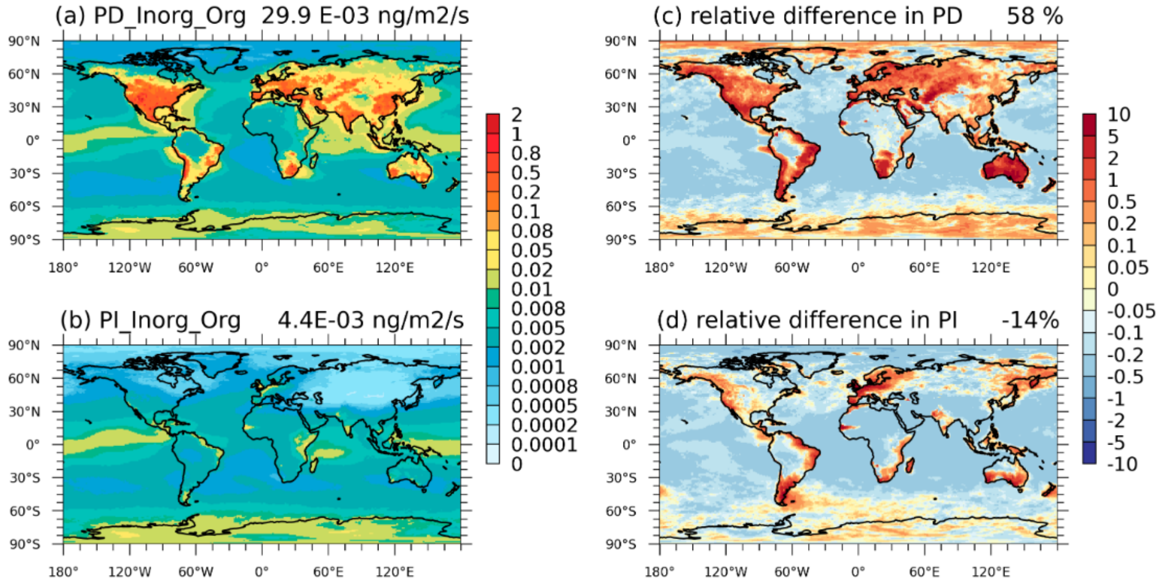

Figure S16. Spatial distribution of the simulated vertically-integrated sulfuric acid nucleation loss rate in (a) PD and (b) PI experiments (unit: ng m^{-2} s^{-1}). The relative difference between Inorg_Org and Inorg in PD and PI experiments is shown in (c) and (d) (unitless). Global mean values are shown on the top right of each figure.

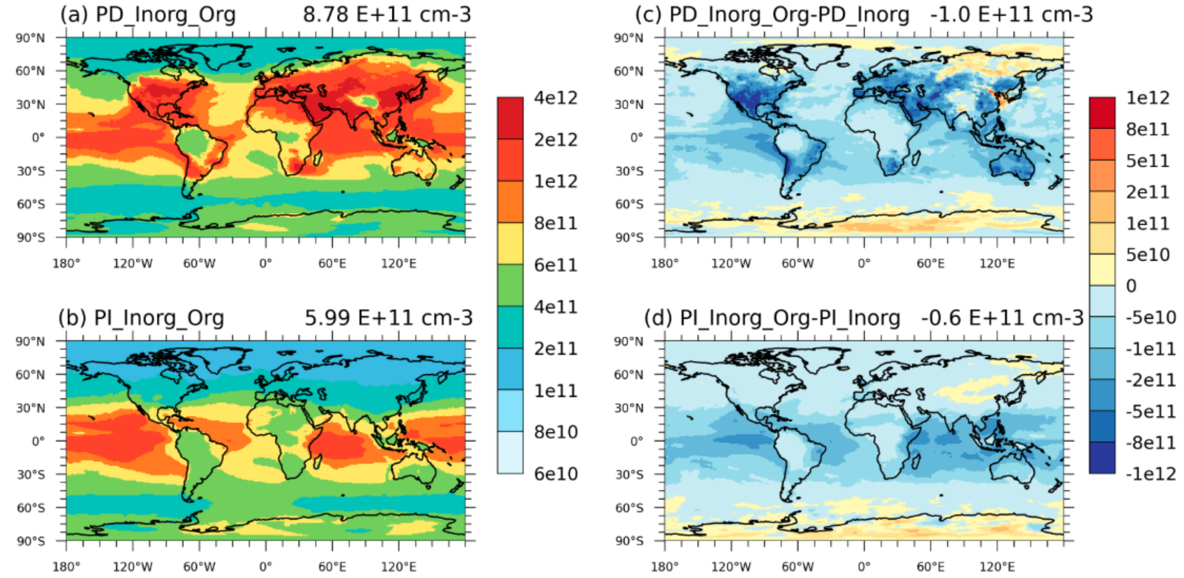

Figure S17. Spatial distribution of the simulated vertically-integrated sulfuric acid concentration in (a) PD and (b) PI experiments (unit: cm^{-2}). The difference between Inorg_Org and Inorg in PD and PI experiments is shown in (c) and (d) (unitless). Global mean values are shown on the top right of each figure.

Major Comment#7: Fig. 1: According to Fig. 1, the improvements of the Shao 2024 organic nucleation scheme (Inorg_Org) is not very significant and distinguishable from the Inorg run. It does not appear to the reader that the data points are significantly close to the 1:1 line or the NMB is largely reduced. Also, reader is curious about the correlation coefficient, which might not show significant improvements either. Please include correlation coefficients for all panels.

Multiple measurements still have predictions several orders of magnitude off from the 1:1 line. Overall many data points are stacked together so it is hard to examine the comparison region by region. From reader's perspective, this figure is not showing the merits of the new nucleation scheme.

Response: Thank you for your constructive feedback. We have revised Figure 1 to use bar plots instead of scatter plots to better compare the performance between the two simulations across different regions (categorized into Marine, Urban, Rural, and Mountain environments), suggested by reviewer#2. The original scatter plot has been moved to the Supplementary Information as Figure S18, with correlation coefficients (R) provided for reference.

We have revised the original description in **Section 3**:

Original Paragraph:

"CCN number concentrations in the Inorg_Org model at 0.1%, 0.2%, 0.5%, and 1% supersaturation (ss) show better agreement with measurements from various locations compared to the Inorg model (Fig. 1). The underestimation of CCN numbers in the Inorg simulation is alleviated by incorporating organic-related NPF, especially over rural and mountainous regions such as Steamboat Springs, Shouxian, and Nainital (Fig. 1), where both nucleation and initial growth rates are dominated by biogenic pathways. However, there remains a slight underestimation of CCN number in Shouxian at all supersaturation levels. This is likely due to the neglect of anthropogenic-derived HOMs to nucleation and growth, which are key NPF mechanisms in rural regions of China. The increase in CCN number due to the addition of organic NPF mechanisms is simulated not only in the locations listed in Table 1 but also on a global scale (see Fig. 7). "In urban regions of Brazil (Manacapuru), the overestimation of CCN numbers at 0.5% and 1% ss in Inorg is exacerbated (Fig. 1). Apart from urban regions like Manacapuru, Brazil, the overestimation also increases over oceanic regions such as Barrow and Graciosa. These overestimations in CCN numbers in the Inorg model are likely related to the overestimation of H_2SO_4 concentration in CAM6-Chem (Shao et al., 2024). Overall, the normalized mean bias (NMB) of CCN numbers at different supersaturation levels decreases from -35% (Inorg) to -24% (Inorg_Org), indicating that the Inorg_Org model provides a more accurate representation of organic contributions for further quantification in Section 3."

Revised Paragraph:

"The underestimation of CCN numbers in the Inorg simulation is alleviated by incorporating organic-related NPF, especially over rural and mountainous regions (Fig. 1), where both nucleation and initial growth rates are dominated by biogenic pathways. The remaining underestimation of CCN in rural regions (Fig. 1) is likely due to the neglect of anthropogenic-derived HOMs, which may play a key role in NPF in these areas. The increase in CCN number due to the addition of organic NPF mechanisms is simulated not only in the locations listed in Table 1 but also on a global scale (see Fig. 7). In urban regions, the overestimation of CCN numbers is exacerbated (Fig. 1). These overestimations in CCN numbers in the Inorg model are likely related to the overestimation of H_2SO_4 concentration in CAM6-Chem (Shao et al., 2024). Overall, the relative bias of CCN numbers at different supersaturation levels decreases from -57% (Inorg) to -45% (Inorg_Org) (Fig. S18), indicating that the Inorg_Org model provides a more accurate representation of organic contributions for further quantification in Section 3."

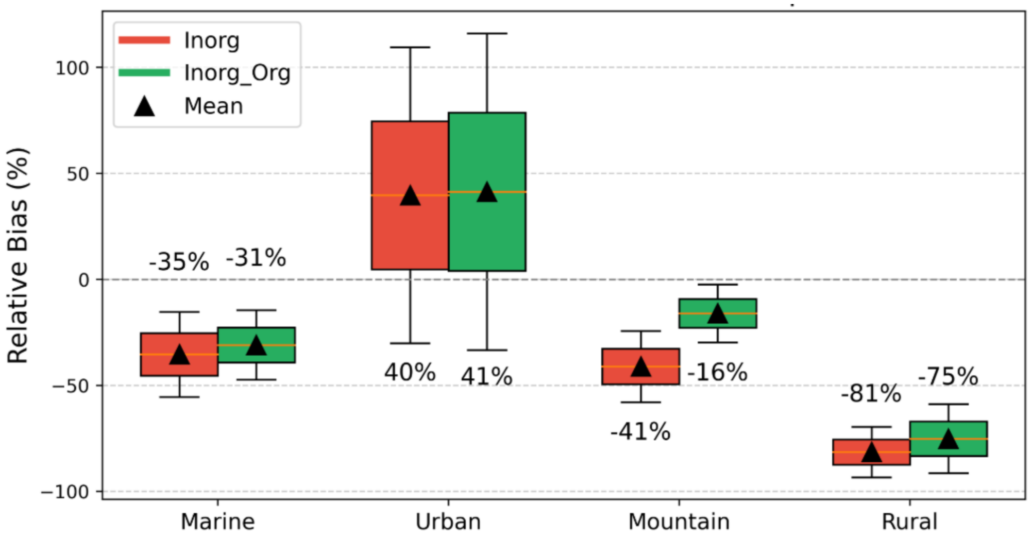

Figure 1. Box plots showing the relative bias (%) between simulated monthly mean and observed median CCN number concentrations across categorized background sites (Marine, Urban, Mountain, Rural). Red and green boxes represent the Inorg and Inorg_Org experiments, respectively. Black triangles indicate the mean relative bias for each category. Numerical values above the boxes denote the corresponding mean normalized mean bias (NMB) for each experiment. Information on the measurement sites is provided in Table 2.

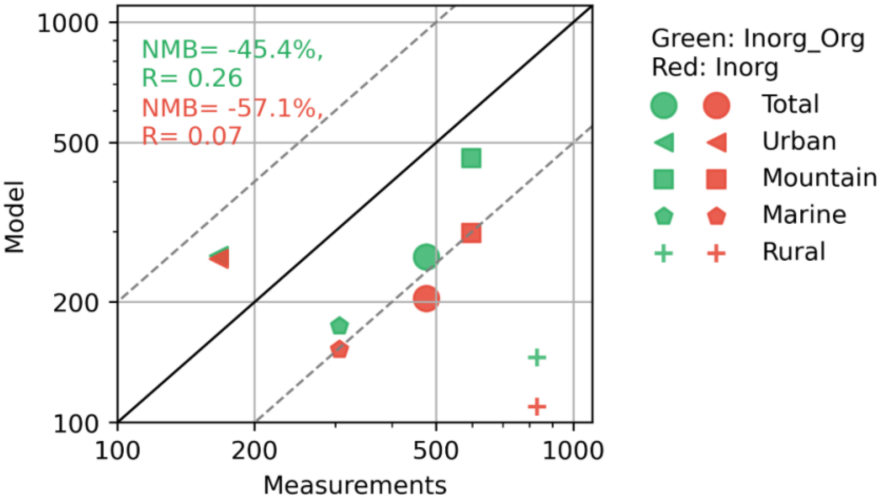

Figure S18. Comparison of simulated monthly mean and observed median CCN number concentrations (unit: cm^{-3}) at categorized background sites. Results from the Inorg_Org experiment are shown in green, and those from the Inorg experiment are shown in red. Information on the measurement sites is provided in Table 2. Normalized mean bias (NMB) and correlation values are indicated in the top-left corner of each panel.

Major Comment#8: Lines 185-186: Why does 0.2% SS in Fig. 1b has much fewer CCN number concentrations than other panels in Fig. 1?

Response: We apologize for the confusion. Please note that the panels in Fig. 1 are not plotted on the same scale, which may give the impression that CCN number concentrations at 0.2% SS are much lower. In general, a lower supersaturation results in fewer particles being activated as CCN, as predicted by Köhler theory, compared to higher supersaturation levels. The revised figure and corresponding description have already been provided in response to the **Major Comment#7.**

Major Comment#9: Lines 194-195: This sentence does not make much sense to reader. Why would a CCN map (Fig. 2) with a much wider area coverage than the coverage of an aerosol burden map (Figs. S11-S12) be due to the slower removal rate of larger aerosol particles? Please rewrite and clarify.

Response: The original text of lines 194-195 is:

"The spatial pattern of change in CCN burden (Fig. 2) is consistent with the change in aerosol burden in the Aitken and accumulation mode (Figs. S11 and S12) but affects much wider areas because of the slower removal rate of larger aerosol particles."

This means the lifetime of CCN (cloud condensation nuclei) is typically longer than that of aerosol particles and is less easily removed (Pierce et al., 2009; Riemer et al., 2009). As a result, CCN can be transported over longer distances and influence a wider area, which explains why the spatial extent of CCN impact is greater than that of aerosol burden.

To avoid further misunderstanding, sentences in Lines 194-195 were modified as (The underlined content is newly added or modified):

"The spatial pattern of change in CCN burden (Fig. 2) is consistent with the change in aerosol burden in the Aitken and accumulation mode (Figs. S11 and S12) but affects much wider areas. ~~because of the slower removal rate of larger aerosol particles~~. Since ultrafine particles (< 50 nm) are quickly lost by coagulation to larger, pre-existing aerosol, CCN typically have a longer atmospheric lifetime and are less efficiently removed than smaller aerosol particles, allowing them to exert influence over wider spatial scales (Pierce et al., 2009; Riemer et al., 2009)."

Major Comment#10: Lines 254-255: Do you have mode-specific values to let readers understand the changes in the indirect forcings for each mode?

Response: Thank you for your valuable comment. Although the model output does not provide mode-specific values for changes in indirect radiative forcing, we have provided supplementary data (Figure S19) on the particle number size distribution (PNSD) by mode. We hope this will help readers better understand the contributions of different aerosol modes to the overall indirect forcing changes.

Although the number concentration of Aitken mode aerosols exhibits substantial changes after incorporating organic NPF (Fig. S19), only particles larger than 50 nm in diameter can typically activate as CCN under ambient supersaturations (Dusek et al., 2006). Therefore, the change in indirect radiative forcing is mainly dominated by the accumulation mode, as it is the only mode showing significant number concentration changes within the CCN-relevant size range (Fig. S19).

The sentence in Line 255 was modified as (The underlined content is newly added or modified):

"The calculated aerosol ERF_{aci} decreases by approximately 0.4 W m^{-2} (corresponding to a 16% reduction) after adding organic NPF mechanisms (Fig. 6b), primarily driven by changes in the number concentration of accumulation mode aerosols (Fig. S19)."

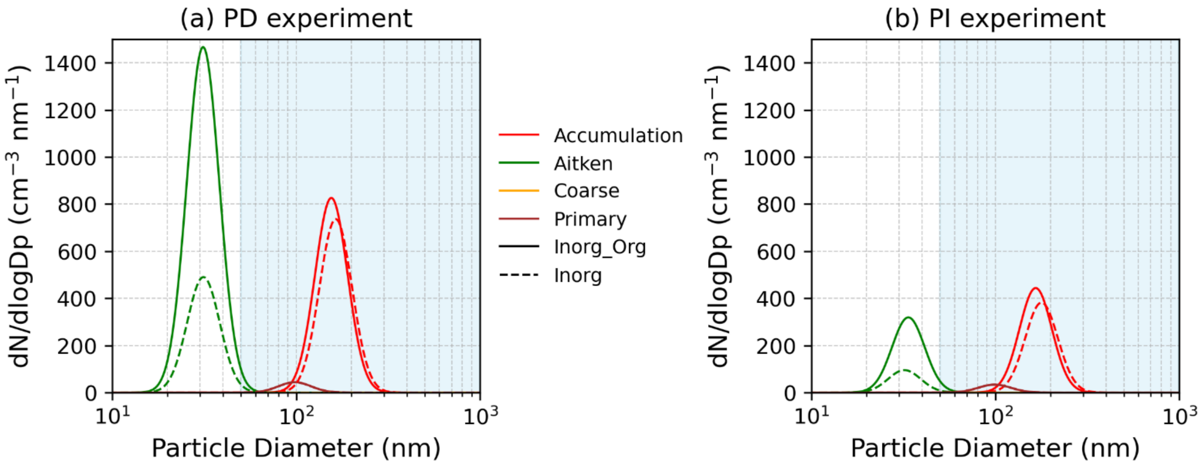

Figure S19. Particle number size distribution (PNSD) by mode in the (a) PD and (b) PI experiments. Solid lines represent the Inorg_Org simulation, and dashed lines represent the Inorg simulation. Each color corresponds to a specific mode: Accumulation (red), Aitken (green), Coarse (orange), and Primary (brown). The shaded blue area highlights particles with diameters larger than 50 nm, which are typically large enough to activate as CCN under ambient supersaturation conditions (Dusek et al., 2006).

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