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#3

General Comments:

The manuscript addresses a significant knowledge gap regarding the role of highly oxygenated organic molecules (HOMs) in new particle formation (NPF) during the preindustrial era, which is critical for establishing accurate baseline conditions for radiative forcing calculations. The implementation of a semi-explicit HOMs chemistry scheme (in CAM6-Chem) represents a significant improvement compared to the simplified fixed-yield approaches. A very interesting finding is that the condensational growth (before Aitken mode particles are formed) rather than nucleation itself is the primary driver of CCN enhancement in preindustrial conditions. The manuscript is well written, with great supporting materials that help the readers to better understand the results. On the other hand, I think the manuscript could be further improved in the following aspects:

Response: We thank the referee for the positive and insightful comments, which indeed help us further improve the manuscript. We have incorporated the necessary revisions to address the comments, and the corresponding line numbers have been indicated in the revised manuscript. Please see the revision and the response for the comments as follows.

Comment#1: The manuscript identifies enhanced growth rates of small particles in the PI atmosphere as the primary driver for increased CCN concentrations and subsequent reduction in ERFaci. Since the impact of new treatment on sub-20nm particle growth under preindustrial (PI) conditions is one of the key findings of this study, it would be beneficial to include more analysis and discussion on this topic in the main text. For example, it would be useful to present figure S13 (panels c,d) and corresponding results for "PI_Inorg" and compare the growth rates between the two cases under PD&PI conditions. If there are model diagnostics of condensation rates by sulfuric acid gas and by organics, it would be valuable to compare them.

Response: Thank you for this valuable suggestion. We have revised the main text to include further discussion on this topic. Specifically, we have moved Figure S13 panels (c, d) into the main manuscript (now shown in Fig. 8) and added the growth rate in "PI_Inorg" to compare the growth rates between the two cases under both PD and PI conditions. Additionally, the condensation rates of H_2SO_4 gas and organics in PD and PI experiments are now added in the supplementary materials (Fig. S14).

Sentences in Lines 283-285 were modified as (The underlined content is newly added):

"Therefore, the greater enhancement of CCN burden in the PI experiment and the reduction in ERF_{aci} are mainly caused by organic condensational growth on sub-20 nm particles (with PD fractional changes of 6% in Inorg_Org and 58% in Inorg; Fig. 7), rather than by organic nucleation. Specifically, after incorporating the organic NPF mechanism, the growth rate of sub-20 nm particles increases more significantly in the PI experiment (0.0083 nm h^{-1}) than in the PD experiment (0.0036 nm h^{-1}) (Fig. 8). This is mainly due to the higher organic sub-20 nm growth rate in PI (0.01 nm h^{-1}) compared to PD (0.006 nm h^{-1})."

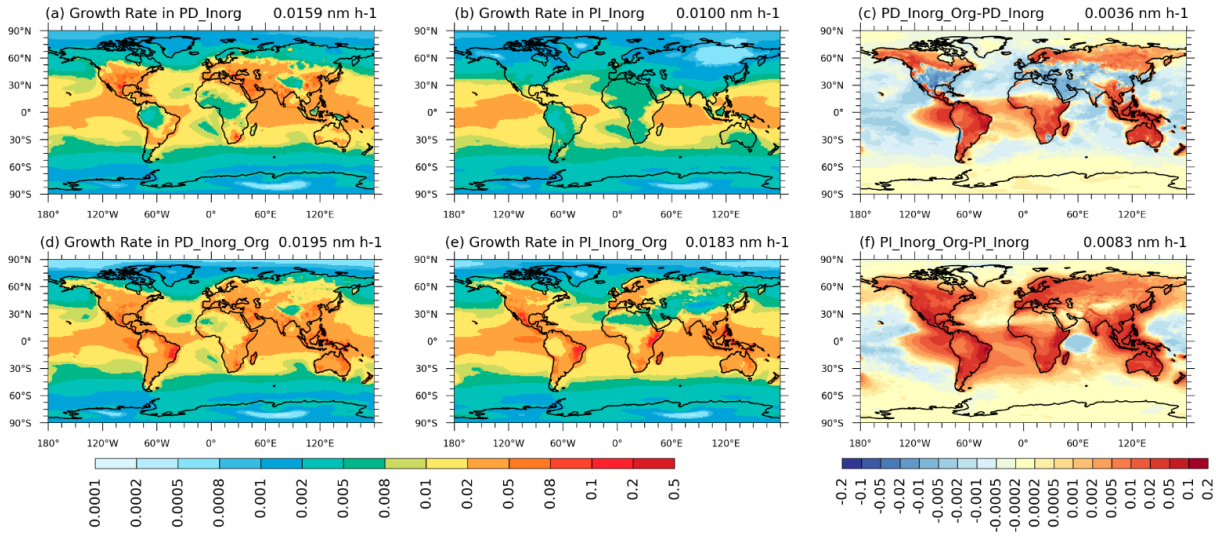

Figure 8. Spatial distribution of the simulated vertically-mean growth rate in (a and d) PD and (b and e) PI experiments. The difference between Inorg_Org and Inorg in PD and PI experiments is shown in (c) and (f) (unit: nm h^{-1}). Global mean values are shown on the top right of each figure.

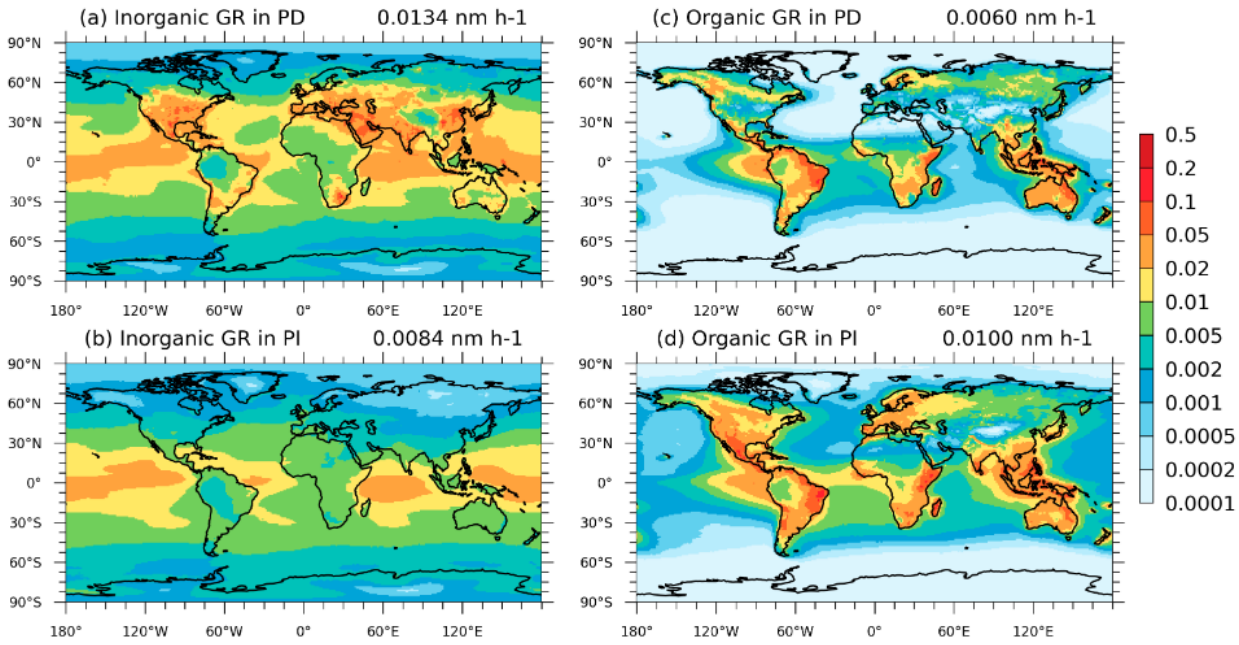

Figure S14. Spatial distribution of the simulated vertically-mean inorganic growth rate (a and b) and organic growth rate (c and d) in PD and PI experiments (unit: nm h^{-1}). Global mean values are shown on the top right of each figure.

Comment#2: The present study discusses the impact of the new nucleation treatment on indirect aerosol effect. It would be more meaningful to also present total effective aerosol forcing changes (either in figure or table), in addition to the decomposed values, since there are often compensating effects between different forcing components.

Response: Thank you for your suggestion. We have added the total effective aerosol forcing and effective radiative forcing due to aerosol-radiation interactions (ERF_{ari}) changes in a table (Table S4) to provide a clearer understanding.

The description of the aerosol forcing in Line 250 of Section 4.2 was modified as (The underlined content is newly added or modified):

"We estimate that the global ERF_{aci} since 1850, after including organic NPF, is -2.18 W m^{-2} (Fig. 6a). 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). The global mean effective radiative forcing due to aerosol-radiation interactions (ERF_{ari}) changes only slightly, from 0.03 W m^{-2} to -0.01 W m^{-2}, a negligible change compared to the total aerosol radiative forcing, which decreases from -2.19 W m^{-2} to -2.64 W m^{-2} (Table S4)."

Table S4. Decomposition of the global aerosol radiative forcing in different experiments (W m^{-2}).

	Inorg_Org (W m^{-2})	Inorg (W m^{-2})	Inorg_Org-Inorg (W m^{-2})
Total effective aerosol forcing	-2.19	-2.64	0.45
Effective radiative forcing due to aerosol-cloud interactions (ERF_{aci})	-2.18	-2.59	0.41
Effective radiative forcing due to aerosol-radiation interactions (ERF_{ari})	0.03	-0.01	0.04

Comment#3: Since the authors claim (line 284 and 316) that the CCN burden change and the indirect aerosol effect are mainly caused by ORGANIC condensational growth on sub-20nm particles, it would be useful to isolate this impact by performing an additional simulation with the organic condensational growth switched off (if it is straightforward). Otherwise, I would suggest stating it as "likely" or removing the emphasis on organics condensation.

Response: We acknowledge that this diagnostic quantity was not initially provided. Due to the significant computational resources required for long-term simulations, we are unable to run an additional set of experiments with organic condensational growth switched off.

We believe that Figure 4 in the main text provides strong support for this conclusion. In Figure 4, the impact of organic nucleation on the total nucleation rate in the PI atmosphere is relatively small, accounting for only 6%. This is mainly due to the lower H_2SO_4 concentrations in the PI atmosphere (Fig. S2), which reduces the H_2SO_4-HOM nucleation rate. In contrast, in the PD atmosphere, higher concentrations of H_2SO_4 and HOMs in the Northern Hemisphere mid-to-high latitudes (Figs. S2 and S3) make organic nucleation more significant, leading to a 39% increase in the total nucleation rate. In contrast to organic nucleation, the impact of organic growth rate on the total growth rate is more significant in the PI atmosphere, reaching 83%, while in the PD atmosphere this impact is only 23%. This is mainly due to significantly higher emissions of organic precursors, such as monoterpenes and isoprene in the PI atmosphere (Fig. S3), and the organic growth rate is only influenced by HOMs concentrations. Therefore, when considering the greater increase in CCN burden in the PI experiment compared to the PD experiment (Fig. 2), the increase in sub-20 nm growth rate plays a more significant role than nucleation rate (Fig. 4 and Fig. S13).

To further support this conclusion, we have calculated the correlation between condensational growth rate on sub-20nm particles with CCN burden. A higher correlation (R ~ 0.7) between organic condensational growth rate and CCN burden, as well as a relatively lower correlation (R ~ 0.4), further suggests that the increase in CCN burden is likely driven by organic

condensational growth. Additionally, the Figure 8 added in Response 1 may provide further support for this point.

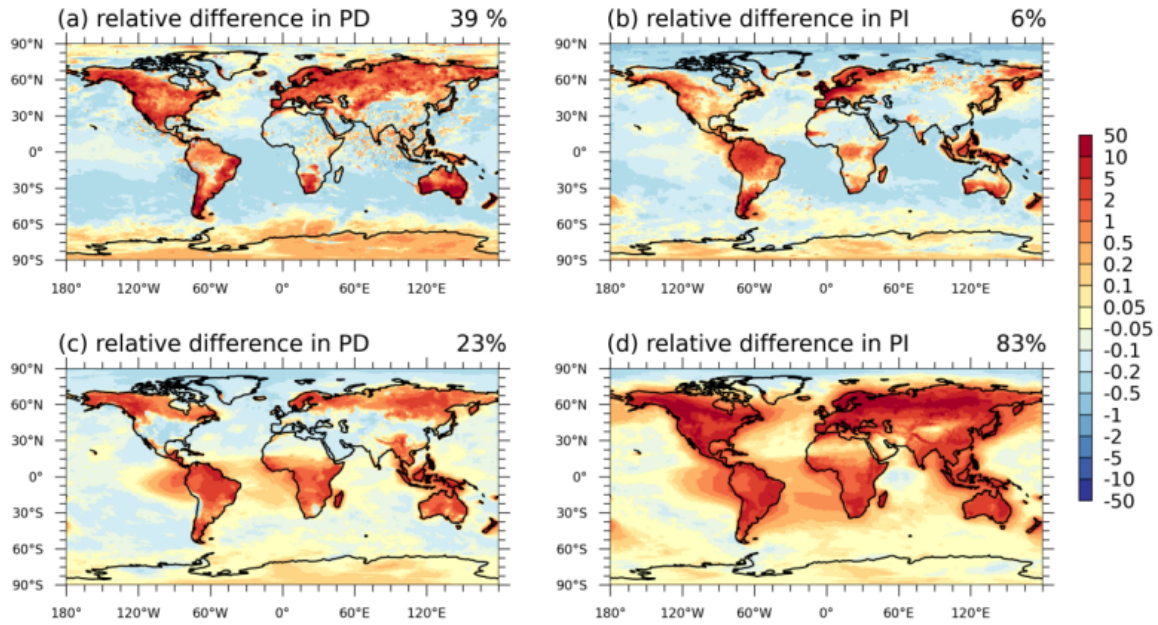

Figure 4. The relative change (unitless) of the simulated (a and b) vertically-integrated nucleation rate ($j_{1.7nm}$, below 15 km) and (c and d) vertically-mean sub-20nm growth rate after adding organic nucleation is shown in PD and PI environments. Global mean values are shown on the top right of each figure.

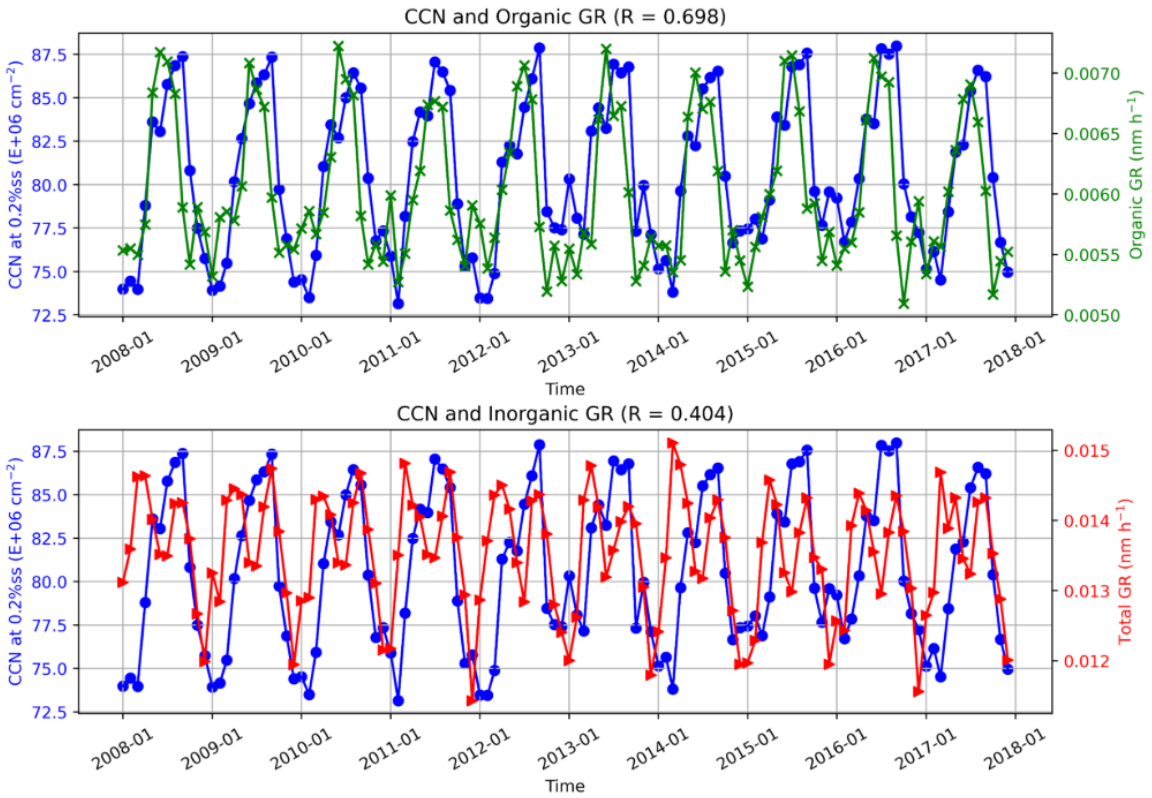

Figure S15. Global mean values of simulated CCN burden at 0.2% supersaturation (blue lines, unit: 10^6 cm^{-2}) and vertically averaged (a) organic and (b) inorganic growth rates (unit: nm h^{-1}) in the

PD_Inorg_Org experiment. Pearson correlation coefficients between growth rates and CCN burden are shown in the titles of each panel.

The description of the aerosol forcing in Line 213 of Section 4.1 were modified as (The underlined content is newly added or modified):

"In both PD and PI experiments, the largest increase in CCN burden (>20% rise in Inorg_Org compared to Inorg) is simulated in the tropical regions (Amazon, central Africa, and Southeast Asia) (Fig. 2). This is attributed to the highest biogenic emissions (Fig. S3) which lead to the greatest increases in both nucleation and growth rates in Inorg_Org (Fig. 4) and the originally low aerosol number before adding organic NPF (i.e., Inorg simulation) in these regions. 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 PD environment (Fig. S2), resulting in higher heteromolecular nucleation rates involving sulfuric acid and organics (Figs. S6 and S7). ~~A detailed discussion of the specific reason is provided in Section 4.2.~~ In contrast to organic nucleation, the impact of organic growth rate on the total growth rate is more significant in the PI atmosphere, reaching 83%, while in the PD atmosphere this impact is only 23%. This is mainly due to significantly higher emissions of organic precursors, such as monoterpenes and isoprene in the PI atmosphere (Fig. S3), and the organic growth rate is only influenced by HOMs concentrations. Therefore, comparing to the increase in the ~1.7 nm nucleation rate, the increase in the sub-20 nm growth rate plays a more significant role in greater increase of CCN burden in PI experiment (Fig. 4 and Fig. S13). The strong correlation (R~0.7) between organic growth rates and CCN burden further supports this point (Fig. S15)."

The description of the aerosol forcing in Line 283 was modified as (The underlined content is newly added or modified):

"Therefore, the greater enhancement of CCN burden in PI experiment and reduction in ERF_{aci} are ~~mainly~~ likely caused by organic condensational growth on sub-20nm particles instead of organic nucleation."

Specific comments:

Figure 3: Is CDNC either vertically-integrated or at the top of low clouds? Which is correct?

Response: CDNC is calculated at the cloud top of low-level clouds. We will update the figure caption to state this explicitly. Figure 3 will be revised as:

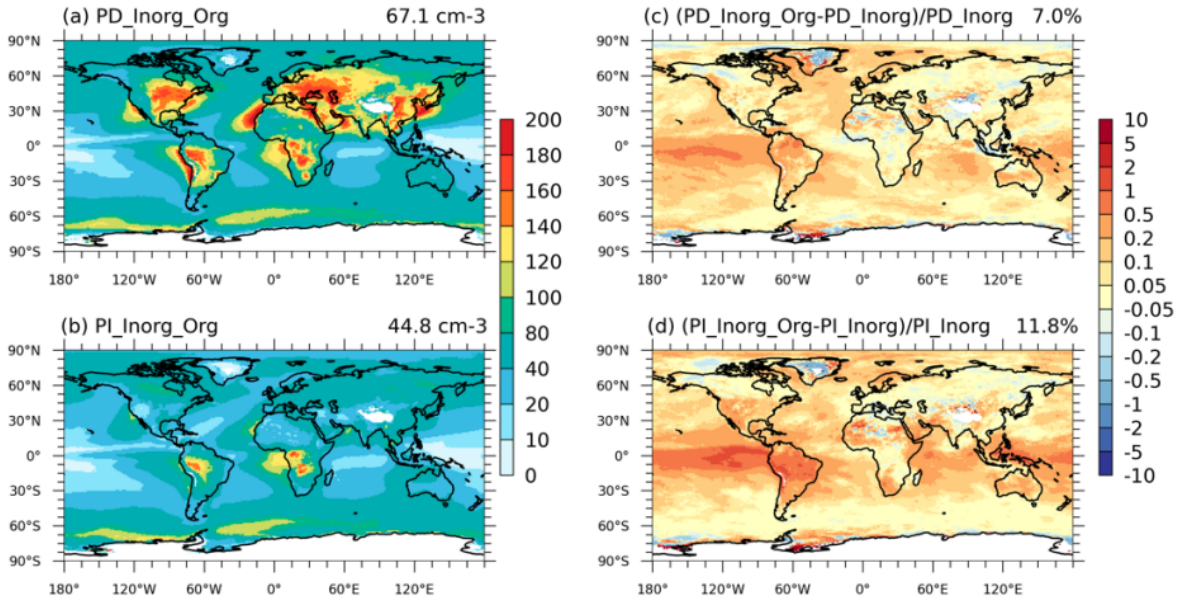

Figure 3. Spatial distribution of the simulated ~~vertically-integrated~~ cloud droplet number concentration (CDNC) at the top of low clouds in (a) PD_Inorg_Org and (b) PI_Inorg_Org (unit: cm^{-3}). The relative change after adding organic NPF ~~is shown~~ in PD and PI environments are shown in (c) and (d). Global mean values are shown on the top right of each figure.

Figure 4: Why are the values over ocean negative in panels a) and b)? Also, the caption is somewhat confusing. I suggest rewriting it.

Response: The main reason for the negative values over the ocean in panels a) and b) is 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 mainly driven by H_2SO_4 (Fig. 7 in Shao et al., 2024), leading to decreased nucleation rate and H_2SO_4 nucleation sink (Fig. S16) over ocean.

In light of this, we have added the following explanation in Line 219 of the main text (The underlined content is newly added or modified):

"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 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 value. Consequently, more H_2SO_4 is consumed over land (Fig. S16), reducing its transport to oceanic regions. As a result, nucleation rates decrease over the ocean in both the PD and PI experiments (Fig. 4)."

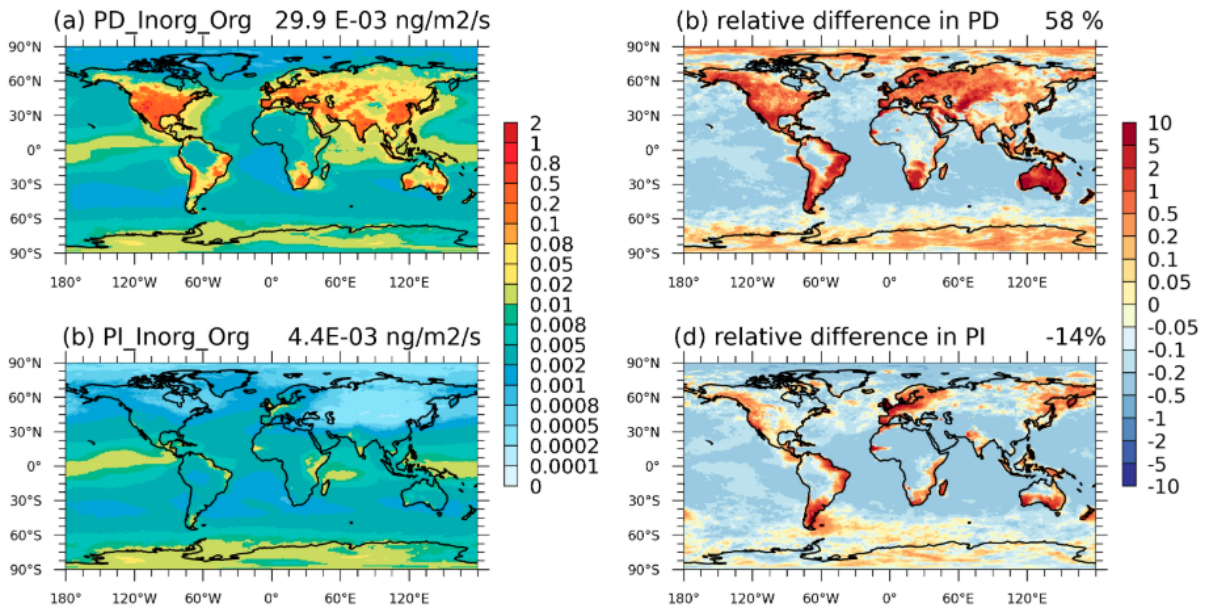

Figure S16. Spatial distribution of the simulated vertically-integrated sulfuric acid nucleation loss rate in (a) PD and (b) PI experiments (unit: ng m^{-2} s^{-1}). The relative difference between Inorg_Org and Inorg in PD and PI experiments are shown in (c) and (d) (unitless). Global mean values are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

The original caption of Figure 4 was revised as below:

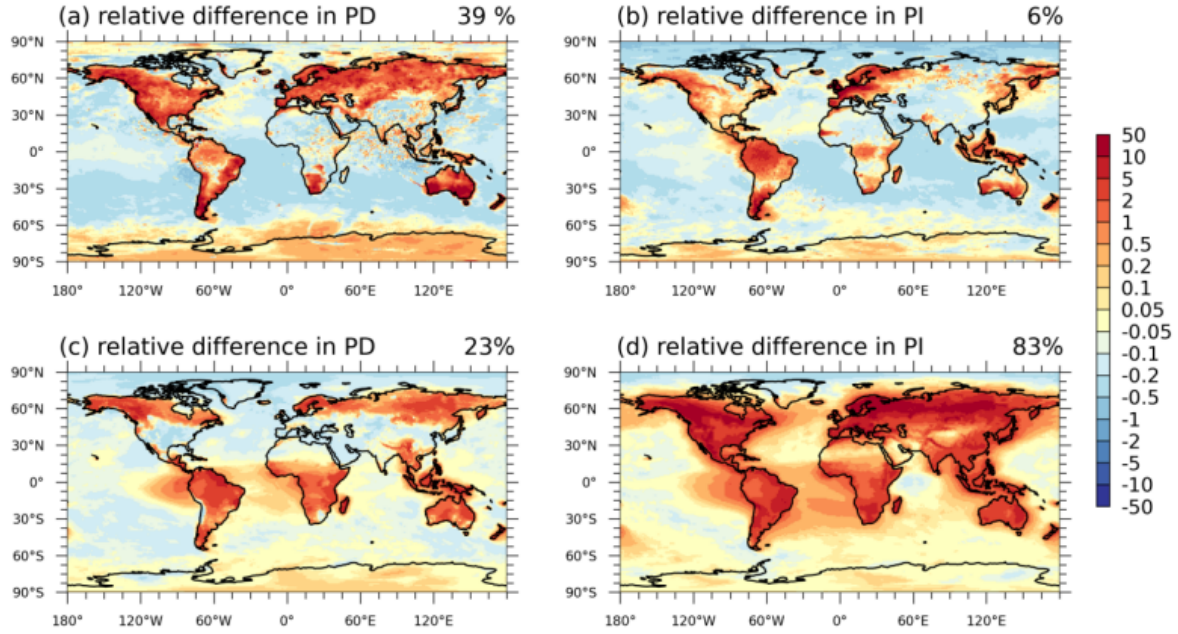

Figure 4. ~~Spatial distribution in (a) PD_Inorg_Org and (b) PI_Inorg_Org (unit: m-2 s-1).~~ The relative change (unitless) of the simulated (a and b) vertically-integrated nucleation rate ($j_{1.7nm}$, below 15 km) and (c and d) vertically-mean sub-20nm growth rate after adding organic nucleation is shown in PD and PI environments. Global mean values are shown on the top right of each figure.

Figure 6: It would be more meaningful to also present total effective aerosol forcing changes (either in figure or table), in addition to the decomposed values. There are often compensating effects between different forcing components.

Response: Thank you for your suggestion. In response to your second major comment, we have made the necessary revisions and included the total effective aerosol forcing changes, in addition to the decomposed values, as recommended.

Line 176: Should be Table 2.

Line 198: "rise" → "rises"

Line 220: "comparing to" → "compared to"

Response: Thank you for your helpful comments. In response to your suggestions, we have made the following revisions in the manuscript:

Line 176: Corrected to "Table 2" as suggested. Line 198: Changed "rise" to "rises". Line 220: Replaced "comparing to" with "compared to".

Line 230-231: Is this really the case in this study? It appears that the accumulation mode aerosol number concentrations have significant changes after considering organic nucleation.

Response: Thank you for your suggestion. We have removed the sentence and added the total effective aerosol forcing and effective radiative forcing due to aerosol-radiation interactions (ERF_{ari}) changes in a table (Table S4) to provide a clearer understanding.

Table S4. Decomposition of the global aerosol radiative forcing in different experiments (W m^{-2}).

	Inorg_Org (W m^{-2})	Inorg (W m^{-2})	Inorg_Org-Inorg (W m^{-2})
Total effective aerosol forcing	-2.19	-2.64	0.45
Effective radiative forcing due to aerosol-cloud interactions (ERF_{aci})	-2.18	-2.59	0.41
Effective radiative forcing due to aerosol-radiation interactions (ERF_{ari})	0.03	-0.01	0.04

The description of the aerosol forcing in Line 230 of Section 4.2 were modified as (The underlined content is newly added or modified):

"The significant increase in CCN number and CDNC in the PI experiment resulting from the inclusion of the organic NPF scheme is likely to reduce the aerosol radiative forcing. ~~The aerosol direct radiative forcing may not be significantly influenced by the NPF mechanism because it is not strongly affected by the aerosol size distribution (Rap et al., 2013). Thus,~~ In this study we ~~only~~ focus on quantifying the effect of including biogenic organic NPF on the indirect aerosol forcing component (ERF_{aci})."

The description of the aerosol forcing in Line 250 of Section 4.2 were modified as (The underlined content is newly added or modified):

" We estimate that the global ERF_{aci} since 1850, after including organic NPF, is -2.18 W m^{-2} (Fig. 6a). 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). The global mean effective radiative forcing due to aerosol-radiation interactions (ERF_{ari}) changes only slightly, from 0.03 W m^{-2} to -0.01 W m^{-2}, a negligible change compared to the total aerosol radiative forcing, which decreases from -2.19 W m^{-2} to -2.64 W m^{-2} (Table S4)."

Line 280: "organics rate" → "organics"

Line 295: Delete "was neglected"

Line 318: "is" → "are"

Line 324: Should be Figure 6 (not 9).

Line 325: "higher increase" → "greater increase"

Response: Thanks. We have made the corresponding revisions in the manuscript based on your suggestions.

Line 337: "with a greater reduction than estimated in previous studies." This appears inconsistent with Gordon et al.'s higher percentage reduction (27%) than this study (16%).

Response: Thank you for pointing out the inconsistency. What we intended to emphasize was the comparison with Zhu et al. (2019) (16% reduction), as Zhu et al. (2019) employed a more advanced chemical mechanism compared to Gordon et al. (2016). To avoid any confusion, we will remove this paragraph.

Reference

Gordon, H., Sengupta, K., Rap, A., Duplissy, J., Frege, C., Williamson, C., Heinritzi, M., Simon, M., Yan, C., Almeida, J., Trostl, J., Nieminen, T., Ortega, I. K., Wagner, R., Dunne, E. M., Adamov, A., Amorim, A., Bernhammer, A. K., Bianchi, F., Breitenlechner, M., Brilke, S., Chen, X. M., Craven, J. S., Dias, A., Ehrhart, S., Fischer, L., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Hakala, J., Hoyle, C. R., Jokinen, T., Junninen, H., Kangasluoma, J., Kim, J., Kirkby, J., Krapf, M., Kurten, A., Laaksonen, A., Lehtipalo, K., Makhmutov, V., Mathot, S., Molteni, U., Monks, S. A., Onnela, A., Perakyla, O., Piel, F., Petaja, T., Praplanh, A. P., Pringle, K. J., Richards, N. A. D., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Scott, C. E., Seinfeldo, J. H., Sharma, S., Sipila, M., Steiner, G., Stozhkov, Y., Stratmann, F., Tome, A., Virtanen, A., Vogel, A. L., Wagner, A. C., Wagner, P. E., Weingartner, E., Wimmer, D., Winkler, P. M., Ye, P. L., Zhang, X., Hansel, A., Dommen, J., Donahue, N. M., Worsnop, D. R., Baltensperger, U., Kulmala, M., Curtius, J., and Carslaw, K. S.: Reduced anthropogenic aerosol radiative forcing caused by biogenic new particle formation, P. Natl. Acad. Sci. USA, 113, 12053-12058, 10.1073/pnas.1602360113, 2016.

Zhu, J., Penner, J. E., Yu, F., Sillman, S., Andreae, M. O., and Coe, H.: Decrease in radiative forcing by organic aerosol nucleation, climate, and land use change, Nat. Commun., 10, 423, 10.1038/s41467-019-08407-7, 2019.