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#2**

**General Comments:**

This is an interesting study. The authors incorporated a HOMs-related chemistry and nucleation scheme into a climate model and explored the influence of the HOMs-derived NPF on CCN formation, along with the ensued changes in radiative forcings caused by ACI. Their results showed that including NPF mechanisms in the model can improve simulations of CCN number concentrations. With the new scheme, the authors found that significantly more CCN are generated from organic NPF in PI than PD, which greatly weakens the effective radiative forcing due to ACI. They argued that the weakened radiative forcings are not caused by altered nucleation rates as proposed by Gordon et al. (2016). Rather, it is primarily driven by the more enhanced sub-20 nm growth rate in PI compared to PD. I enjoyed reading this paper, which is very well organized and easy to follow. But there are a few grammatical errors that should be corrected. I have listed some below, but not exhaustive. This work demonstrates the important role of biogenic NPF in CCN formation especially in PI, which enhances our understanding of aerosol emissions and associated nucleation processes in radiative forcings and might help reduce the uncertainties of ACI among climate models in the future. Therefore, I believe this paper will be well-suited for publication in ACP if a few minor issues are addressed.

**Response**: We sincerely thank the referee for the positive and thoughtful comments, which indeed help us further improve the manuscript. We have carefully reviewed the manuscript and corrected the grammatical issues you noted, along with other minor edits. Please see the revision and the response for the comments as follows.

**Comment#1:** An important finding of this study is that the greater increase in CCN in PI than PD is attributed to organic condensational growth on sub-20 nm particles. I wonder whether the authors have examined the full CCN budget to rule out the impact of other contributing processes. For example, the sinks of CCN via precipitation scavenging might also play an important role.

**Response**: While the initial model output did not include these variables, we have examined all aerosol types that could be activated into CCN (including $SO_4$, SOA, POM, Sea Salt) and separately calculated their dry and wet deposition fluxes in both the PD and PI experiments for reference (Table S5). Compared to the growth rate, the PD fractional change in the dry and wet deposition fluxes did not show significant changes after adding organic NPF (values of "Inorg_Org - Inorg" in Table S5).

The sentence in Line 221 was modified as (The underlined content is newly added or modified):

"Therefore, compared 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 the greater increase of CCN burden in the PI experiment (Fig. 4 and Fig. S13). Other components of the CCN budget show no substantial changes (Table S5), further reinforcing the dominant role of condensational growth."

**Table S5**. Global mean of dry and wet deposition fluxes of different aerosols (including sulfate, secondary organic aerosol, primary organic matter and sea salt) in PD_Inorg_Org experiments. The PD fractional change ((PD-PI)/PI) of these variables in Inorg_Org and Inorg experiments are also shown. These aerosol sinks serve as proxies for CCN removal processes.

| | Aerosol Type | **Value** | **PD Fractional Change** | | |
|---|---|---|---|---|---|
| | | PD_Inorg_Org ($ng\ m^{-2}\ s^{-1}$) | Inorg_Org | Inorg | Inorg_Org-Inorg |
| dry deposition flux | $SO_4$ | 0.82 | 25.4% | 25.7% | -0.3% |
| | SOA | 0.08 | 30.6% | 32.2% | -1.6% |
| | POM | 0.04 | 54.9% | 32.2% | -1.6% |
| | Sea Salt | 2.29 | -2.4% | -2.5% | 0.1% |
| wet deposition flux | $SO_4$ | 10.62 | 31.2% | 30.1% | 1.1% |
| | SOA | 0.78 | 49.7% | 48% | 1.7% |
| | POM | 0.78 | 74.5% | 73.9% | 0.7% |
| | Sea Salt | 32.60 | -0.1% | 1.1% | -1.2% |

**Comment#2:** To minimize the influence of meteorological fields on the results, the authors nudged T, winds, and others to MERRA2 reanalysis in the short-term simulations. But the methodology of the nudging experiment is not clearly described, such as what nudging time scale and temporal resolution of MERRA2 are used. A brief examination of simulated winds fields against MERRA2 would also be helpful to strength confidence in the nudging approach.

**Response**: We have included a clear explanation of the nudging methodology and supplemented this with a comparison between the simulated meteorological fields and MERRA2 reanalysis data in the supplementary materials (Fig. S20).

The sentence in Line 143 was modified as (The <u>underlined</u> content is newly added):

"In order to compare simulated CCN with measurements, several short-term simulations were performed, in which meteorological fields (temperature and wind profiles, surface pressure, surface stress, surface heat and moisture fluxes) were nudged toward Modern-Era Retrospective analysis for Research and Applications (MERRA2) reanalysis <u>with a relaxation timescale of 6 hr</u> (Kooperman et al., 2012). <u>Meteorological fields are nudged towards the MERRA2 every 0.5 h, which is the same as the physics timestep of the model (Lamarque et al., 2017). The simulated meteorological fields and their deviations from MERRA2 reanalysis are presented in Figure S20.</u>"

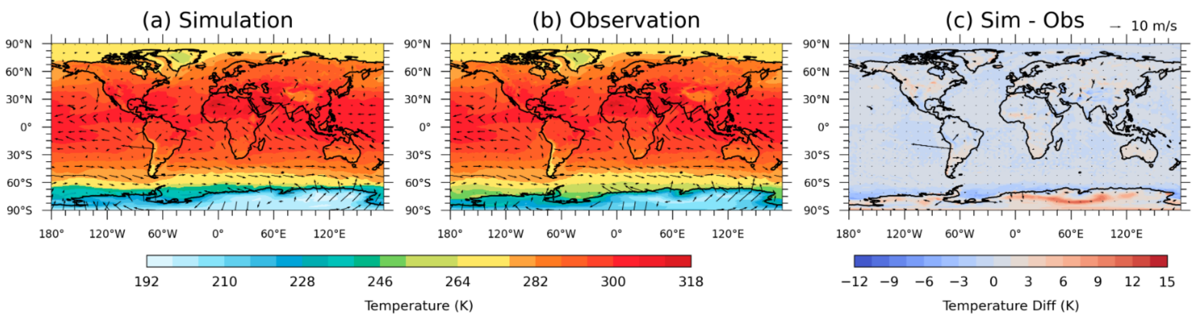


**Figure S20.** Spatial distribution of (a) simulated and (b) observed temperature (shaded, unit: K) and wind speed (arrows, unit: m $s^{-1}$). Panel (c) shows the difference in temperature between simulation and observation.

**Comment#3:** Since the primary goal of Figure 1 is to compare the bias errors of two simulation experiments against observations, rather than to examine correlations, I strongly recommend replacing the scatter plots with bar plots. The current scatter plots do not effectively convey the contrast between the two experiments. Instead, bar plots would allow a clearer demonstration of the simulation errors for each experiment across different regions. To enhance interpretability, the regions could be categorized into Marine, Urban, and Mountain environments, using distinct colors for each category. Additionally, to examine whether the improvement in CCN simulations by Inorg_Org is statistically significant, error bars should be added also.

**Response**: Thank you for the thoughtful suggestion. As recommended, 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). The original scatter plot has been moved to the supplementary material as Figure S18.

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."

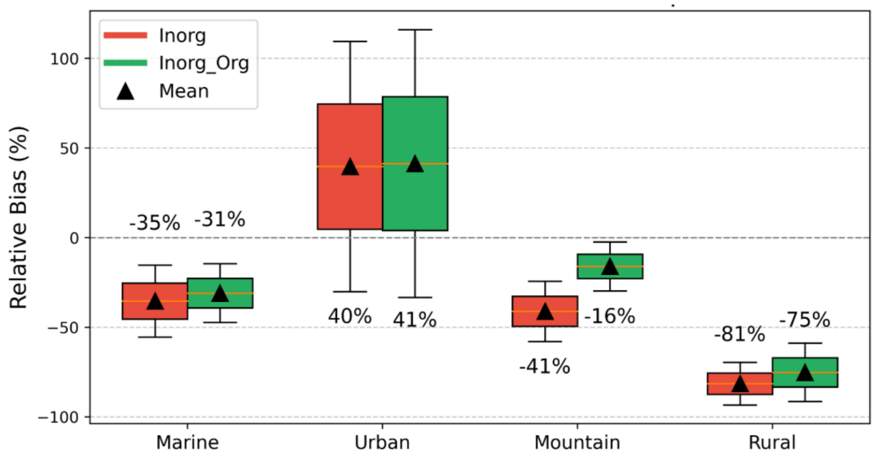


**Figure 1.** Box plots showing the relative bias (%) between simulated monthly mean and observed median CCN number concentrations across categorized background sites (Marine, Urban, Mountain, Rural). Red and green boxes represent the Inorg and Inorg_Org experiments, respectively. Black triangles indicate 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.

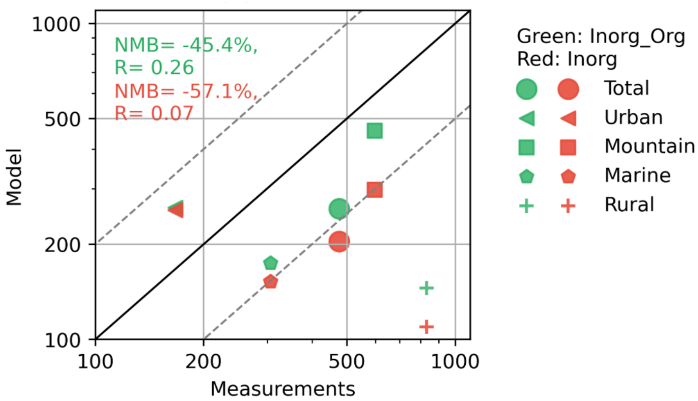


**Figure S18.** Comparison of simulated monthly mean and observed median CCN number concentrations (unit: $cm^{-3}$) at categorized background sites. Results from the Inorg_Org experiment are shown in green, and those from the Inorg experiment are shown in red. Information on the measurement sites is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

**Specific comments:**

L30-32: The authors may consider removing the sentences "while the greater … nucleation rates involving sulfuric acid and organics". The current sentences look confusing here and disrupt the 2 reading flow. Alternatively, if the authors wish to retain these sentences, they need to rephrase these sentences to more clearly align with the argument proposed by Gordon et al. (2016).

**Response**: This sentence has been deleted in the revised manuscript to eliminate confusion. Also, to improve clarity and better align with the argument proposed by Gordon et al. (2016), the sentence in Lines 28-29 was 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 30 rate ($j_{1.7nm}$)~~. 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 ($j_{1.7nm}$) drives the reduction."

L57: "are they" to "they are"

L59: "stringent" to "rigorous"

L213: "simulated" to "found"

L215: "a low" to "the originally low"

L221: "in PI" to "in the PI"

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

L171-172: It is difficult to discern the regional variation in bias from Figure 1.

**Response:** I have made the revision to Figure 1 based on Comment #3. We hope this has improved the figure's readability.

L224: Add description for panels (c) and (d)

**Response:** Sorry for the missing information. The caption for Figure 4 has been updated to include descriptions for panels (c) and (d) (The underlined content is newly added):

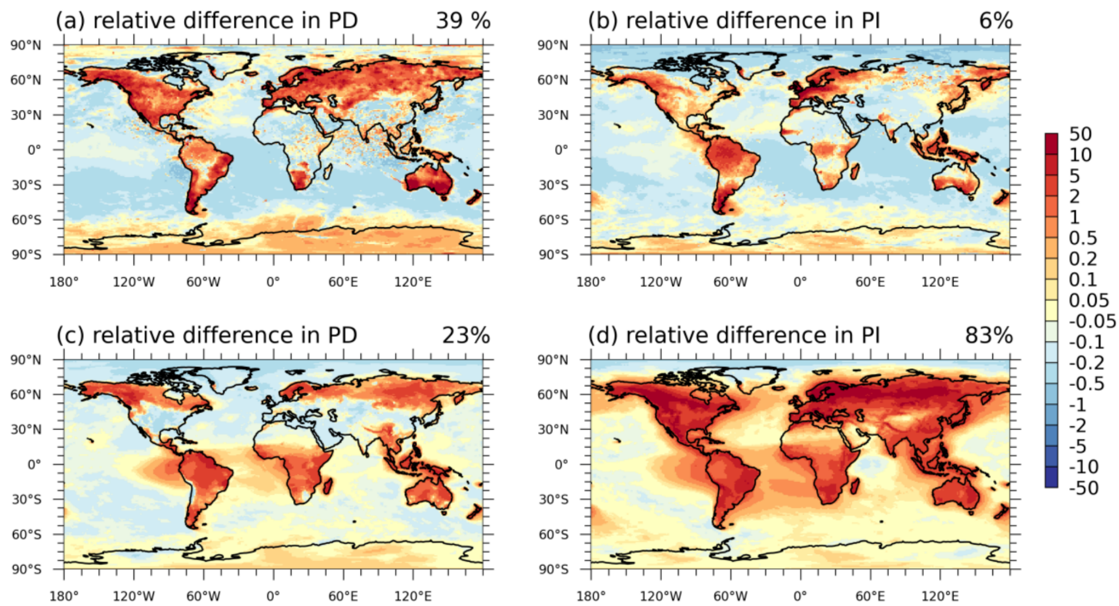


Figure 4. ~~Spatial distribution in (a) PD_Inorg_Org and (b) PI_Inorg_Org (unit: m-2 s-1).~~ The relative change (unitless) of the simulated (a and b) vertically-integrated nucleation rate ($j_{1.7nm}$, below 15 km) and (c and d) vertically-mean sub-20nm growth rate after adding organic nucleation is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

L229: CCN number concentration?

**Response:** The sentence in Line 229 was modified as (The underlined content is newly added or modified):

"The significant increase in CCN ~~number~~ burden (Fig. 2) and CDNC (Fig. 3) in the PI experiment resulting from the inclusion of the organic NPF scheme is likely to reduce the aerosol radiative forcing. "

L230: But adding the NPF mechanism would increase aerosol burden, and thus change the direct radiative forcing, although the size distribution might not change.

**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.

**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 was 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 ~~only~~ 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 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)."

L278: "in PI" to "in the PI"

L278: "leading" to "leads"

L281: "compared to PI" to "compared to the PI"

L284: "in PI" to "in the PI"

L295: remove "was neglected"

**Response:** Thank you for your suggestions. We have made these revisions.

L312: Be specific about backgrounds

**Response:** The sentence in Lines 312 was modified as (The underlined content is newly added or modified):

"After incorporating organic NPF scheme with state-of-the-art chemical mechanisms for biogenic HOMs into CAM6-Chem, the simulated CCN numbers agree better with measurements across different backgrounds (including mountain, rural, and marine) (Fig. 1).”

L315-320: To be clearer, the authors should first clarify how organic nucleation changes are responsible for greater enhancement in PI’s CCN burden in previous studies or Gordon et al. (2016). They can then highlight how their findings differ from those earlier results.

**Response**: Thank you for the suggestion. The sentence in Lines 315 was modified as (The underlined content is newly added or modified):

“After incorporating organic NPF scheme with state-of-the-art chemical mechanisms for biogenic HOMs into CAM6-Chem, the simulated CCN numbers agree better with measurements across different backgrounds (Fig. 1). Globally, the inclusion of organic-related NPF processes results in a 39% increase in CCN burden in the PI experiment and an 18% increase in the PD experiment. Similarly, cloud droplet number concentration (CDNC) at the top of low clouds in the Inorg_Org simulation rises by 12% in the PI experiment but only by 7% in the PD experiment. The greater enhancement of CCN burden in the PI experiment is primarily driven by organic condensational growth on sub-20 nm particles, rather than organic nucleation. We noted that previous studies (Zhu et al., 2019; Gordon et al., 2016) attributed the greater enhancement of CCN burden in the PI experiment to organic nucleation, which is likely due to an overestimation of the organic nucleation rate by assuming uniform volatility among all organic nucleating species. In our study, only accretion products generated through self- and cross-reactions of biogenic radicals are allowed to contribute to pure organic nucleation, making heteromolecular nucleation ($J_{SA\text{-}Org}$) the dominant nucleation pathway. Higher $H_2SO_4$ concentrations in the PD environment further enhance nucleation rates compared to the PI atmosphere. ~~The main reason is that sulfuric acid ($H_2SO_4$) concentrations are significantly higher in the PD environment (Fig. S2), leading to higher heteromolecular nucleation rates of sulfuric acid and organics ($J_{SA\text{-}Org}$), which is the largest contributors to organic nucleation in most regions.~~ In contrast, HOM concentrations are higher in the PI atmosphere (Fig. S3) leading to a much greater condensation of organics on sub-20 nm particles in PI experiments.”

L320: add a period at the end of the sentence.

L336: remove “the”

L340: remove the extra period

**Response**: Thank you for your suggestions. The corresponding revisions have been made in the manuscript.

L328-330: Rephase the sentences to emphasize "although our methods improve the simulations of CCN burdens"

**Response**: The sentence in Lines 328 was modified as (The underlined content is newly added or modified):

"Although we improve the simulations of CCN numbers by utilizing ~~utilized~~ explicit chemical reactions to replace the traditional fixed yield method ~~for simulating biogenic HOMs concentrations~~, further studies are needed to better align simulated HOM concentrations with widespread measurements."

**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.