

We thank the editor and referees for their thoughtful and constructive comments. Please find below detailed responses to each comment or question, including notations of improvements to the manuscript. Editor and referee comments are in blue fonts. Changes and improvements to the manuscript are shown in underlined text.

Editor comments:

Many thanks for revising your manuscript. Referee #1 is satisfied with the changes.

However, referee #2 has some general remaining comments. Please take a look at their report and address the comments.

Please see response to comments of referee #2.

In addition, building on these comments, I would like you to further revise the following aspects:

1) Abstract

The abstract is very long. Please make it more concise. We recommend a length of 300 words while addressing the following points:

- The topic of the article and why it is important
- The status of scientific understanding
- The gap in knowledge being addressed
- The objectives, questions or hypotheses of the study
- The approach
- The main results with important quantitative information if appropriate
- The importance and implications of the results

In particular, point 3 should be added more clearly (what is the hypothesis?), whereas points 5 and 6 are addressed in a lot of detail.

We thank the editor for the suggestion. We have modified the abstract accordingly to address the points suggested above. The abstract is also more concise now (below 300 words). The revised abstract now reads:

Nucleation in the free troposphere (FT) and subsequent growth of new particles represent a globally important source of cloud condensation nuclei (CCN). Whereas new particle formation (NPF) has been shown to occur frequently in the upper troposphere over tropical oceans, there have been few studies of NPF at lower altitudes. In addition, the impact of urban emissions and biomass burning on the NPF in tropical marine FT remains poorly understood. In this study, we examine NPF in the lower and mid troposphere (3-8.5 km) over tropical ocean and coastal region using airborne measurements during the recent Cloud, Aerosol and Monsoon Processes Philippines Experiment (CAMP²Ex). NPF was mostly observed above 5.5 km and coincided with elevated relative humidity (RH) and reduced condensation sink (CS), suggesting that NPF occurs in convective cloud outflow. The frequency of NPF increases with altitude, reaching ~50% above 8 km. An abrupt decrease in NPF frequency coincides with early monsoon

transition, and is attributed to increased CS resulting from reduced convective activity and more frequent transport of aged urban plumes. Surprisingly, a large fraction of NPF events in background air were observed in the early morning, and the NPF is likely made possible by very low CS despite low actinic flux. Convectively detrained biomass burning plume and fresh urban emissions enhance NPF as a result of elevated precursor concentrations and scavenging of pre-existing particles. In contrast, NPF is suppressed in aged urban plumes where the reactive precursors are mostly consumed while CS remain relatively high. This study shows strong impact of urban and biomass burning emissions on the NPF in tropical marine FT. The results also illustrate the competing influences of different variables and interactions among anthropogenic emissions, convective clouds, and meteorology, which lead to NPF under a variety of conditions in tropical marine environment.

2) Comparison to previous studies

Please note the journal scope statement that articles with a local focus must clearly explain how the results extend and compare with current knowledge. While I find several statement that you find similar features as in previous studies, I am missing a discussion on your findings extend current knowledge. Please either add brief discussions to the various subsections in section 4 or dedicate a separate section to such a discussion.

Alternatively – instead of adding such a detailed discussion – you may want to consider recategorization of your manuscript to a Measurement Report for which expects conclusions of more limited scope than in research articles.

Following the editor's suggestion, we have added discussions on findings that extend current knowledge. For example, we show that unlike regional NPF in the boundary layer that mostly occurs around noontime, in the FT over tropical oceans, strong radiation is not a necessary condition for NPF and a large fraction of the background NPF occurs under very low CS condition in the early morning when radiation and actinic flux are low. The prevalence of very low CS condition in the early morning is due to a combination of wet scavenging and less convection overnight. In addition, few direct measurements of NPF in biomass burning plumes have been reported. In this study, we found detrained biomass burning plumes strongly enhance NPF in the outflow region of convective clouds once existing particles are efficiently removed by wet scavenging. We also added a subsection discussing the impact of the urban emissions on NPF in tropical marine free troposphere:

Impact of urban emissions on NPF in tropical marine FT is poorly understood as previous measurements were mostly carried out under pristine conditions. The analyses presented above show that depending on the age and altitude, urban emissions can have different effects on NPF. Above 5.5 km (i.e., approximately the freezing level), convectively detrained fresh urban plumes have low CS due to efficient wet scavenging of existing particles, and elevated precursor concentrations contribute to and enhance NPF under the low CS condition in the outflow regions. At lower altitudes (i.e., below freezing level), NPF takes place in detrained fresh urban plumes with higher CS compared to the background. The higher CS is likely due to less efficient wet scavenging at these lower altitudes. High concentrations of precursors from the fresh urban emissions likely made these NPF possible despite relatively high CS. The species participating in these NPF events may include sulfuric acid and amines (Yao et al., 2018). Future measurements, including the precursors and nucleating species, are needed to elucidate the nucleation

mechanisms in the air masses influenced by fresh urban emissions. In aged urban plumes over 7 km, reactive precursors were mostly consumed during the long-range transport from East Asia, while CS remained relatively high. As a result, the aged urban plumes tend to inhibit NPF instead of promoting it as in the case of fresh urban emissions.

3) Conclusions

Currently, your last section is entitled 'Summary' and I think this describes its content well. However, we would expect a balanced section that also includes 'conclusions'. Please add more text on the limitations, novelties and implications of your findings to the last section that should be renamed 'Summary and Conclusions' or only the latter if you decide to shorten the current text.

We have modified the last section accordingly and renamed it as "Summary and conclusions". It now reads:

5 Summary and conclusions

In this study, we examine NPF in the tropical marine FT in the altitude range of 3-8.5 km using airborne measurements collected during the CAMP²Ex campaign. NPF events were classified based on air mass types, including background, biomass burning-influenced, and urban-influenced. The features of key variables, including RH, CS, UV irradiance as well as concentrations of trace gases are presented for different NPF types and over different altitude ranges. No newly formed particles were observed below 3 km, and NPF was rare and mostly observed in urban-influenced air between 3 and 5.5 km. Vast majority of the NPF events were observed above 5.5 km in air that was processed by convective clouds and with low CS. The frequency of NPF increases with altitude, reaching about 50% at 8 km. There is a drastic decrease in NPF frequency from the southwest monsoon to the monsoon transition period, which is attributed to the increased CS resulting from decreased convective activity (i.e., less efficient removal of existing particles) and more frequent transport of aged urban pollution associated with altered meteorological conditions.

Two different types of NPF in background air were observed in the vicinity of convective clouds. One type was observed under the condition of strong UV irradiance around noontime as in previous studies. In contrast, the second type occurred in the early morning with some of lowest CS observed during CAMP²Ex. The very low CS is attributed to a combination of wet-scavenging and less convection (i.e., reduced vertical transport of aerosol particles from near surface to the FT) over night, and it likely makes the second type of background NPF possible despite low UV irradiance and actinic flux.

NPF was observed in BB-influenced air at altitudes of ~ 6.7 km. Convectively detrained biomass burning plume enhances NPF because of elevated precursor concentrations and efficient scavenging of pre-existing particles. The effect of urban emissions on NPF depends on the age of the urban plume and altitude. Newly formed particles in air masses influenced by fresh urban emissions were observed under the condition of low CS in the outflow regions at altitudes between 5.5 and 6.5 km. The NPF was promoted by elevated concentrations of precursors from the fresh urban emissions. At lower altitudes (i.e., below freezing level), a small number of NPF events were observed in detrained fresh urban plumes with higher CS compared to the background. High concentrations of precursors from the fresh urban emissions

likely made these NPF possible despite relatively high CS. Above 7 km, NPF was observed when the background humid air was lifted by convective clouds and mixed into the aged urban plumes. The reactive precursors in the aged urban plumes are mostly consumed during the long-range transport from East Asia, while CS remains relatively high. As a result, the aged urban plumes inhibit NPF instead of promoting it as is the case for the fresh urban emissions.

This study highlights the competing influences of different variables and interactions among anthropogenic emissions, convective clouds, and meteorology, which lead to NPF under a variety of conditions and altitudes. Most earlier studies found that the NPF typically occurs under the conditions of strong solar radiation around noontime. Here we show NPF can occur in the FT with very low CS in the early morning, despite low actinic flux. Depending on their age, urban emissions can either enhance or inhibit NPF in tropical marine FT. Biomass burning plumes strongly enhance NPF in the outflow region of convective clouds once existing particles are efficiently removed by wet scavenging. Due to the lack of measurements of precursors, the nucleation pathways of NPF in different air mass types remain unclear and should be examined in future studies. The impact of urban and biomass burning emissions on NPF, and the subsequent formation of CCN will also need to be examined in the future by combining field observations and model simulations.

Comments of referee #2:

Measurements: there is not much said about aerosol measurements: inlet, losses, characteristics of the sampling system, conditions or pretreatment like drying and so on. Please give some more details!

We thank the referee for the suggestion. We have modified the second paragraph of Sect. 2.1 by adding details of aerosol measurements and inlet. It now reads:

The measurements examined in this study include aerosol properties, carbon monoxide (CO), methane (CH₄) and ozone (O₃) mixing ratios, meteorological parameters, and radiation (see Table 1 for details). Ambient aerosol was sampled by using a “Clarke” style forward facing shrouded solid diffuser that was operated iso-kinetically (Mcnaughton et al., 2007). Two condensation particle counters (CPCs, Model 3756 and 3772, TSI Inc.) measured the total number concentrations of particles nominally larger than ~3 and ~10 nm ($N_{>3\text{ nm}}$ and $N_{>10\text{ nm}}$), respectively. An additional CPC (TSI Model 3772) sampled downstream of a thermal denuder operated at 350 °C and provided non-volatile particle number concentration (nonvolatile $N_{>10\text{ nm}}$). Aerosol size distributions were characterized by a fast integrated mobility spectrometer (FIMS, 10-600 nm; Wang et al., 2017a; Wang et al., 2017b; Wang et al., 2018) and a laser aerosol spectrometer (LAS, Model 3340, TSI Inc., 100-3000 nm). The aerosol samples measured by FIMS and LAS were dried both actively by Nafion driers and passively due to higher aircraft cabin temperature than the ambient. Size distributions provided by LAS were size-corrected assuming a particle refractive index of ammonium sulfate (Moore et al., 2021).

Are data from the CPC after the TD used in this study? To my impression, not all data from table 1 are used in the study. They might be taken out or it should be mentioned, that they are not used here.

Yes, the concentration of non-volatile particles (non-volatile $N_{>10\text{ nm}}$) is used in Sect. 4.3.1 and Fig. 6.

Section 3: What do you mean by ‘incipient particles’? A particular size range? Why do you think, they will be generally removed in clouds? Since you have the measurements, can you estimate how effective that process will be?

Incipient particles refer to the stable clusters from nucleation of gas phase species and typically have diameters between 1-2 nm. This is now clarified in the text. Given the small size of the incipient particles, they are efficiently removed by cloud droplets through coagulation. For example, in a cloud with droplet diameter of 10 μm and a number concentration of 200 cm^{-3} , the e-folding lifetime of 1.5 nm particles is about 90 seconds.

In the first review, I asked for more interpretation in this section, this could be improved, particularly in 3.1. and 3.2.

Please be more precise with height ranges: Sometimes it is said > 6 km, but it is meant between 3 and 6 km. Do not forget the lowest 3 km.

We added more interpretation in Section 3. For example:

“As CS is largely independent of altitude above 5.5 km (Fig. S2), the strong increase of NPF frequency is likely due to lower temperature and higher galactic cosmic rays ionization rate at higher altitudes, at least partially (Kazil et al., 2006).”

“No NPF events were observed from RF12 through RF17, despite the flight tracks overlap with the earlier flights in terms of location and altitude range. This abrupt decrease in NPF frequency coincides with the early monsoon transition starting on 20 September (Hilario et al., 2021), indicating a strong impact of synoptic condition on NPF in this region...”

“The NPF frequency during the MT is lower than that during SWM at most altitudes above 5.5 km (Fig. S3). The decrease of NPF frequency during the MT is likely due to increased CS (Fig. S3b), which may be a result of reduced convective activity as indicated by lower RH (Fig. S3c) and thus reduced wet scavenging. The more frequent long range transport of aged urban plumes may also contribute to the elevated CS during the MT (Hilario et al., 2021).”

We would also like to point out that Sections 3.1 and 3.2 describe general statistics of NPF events observed during CAMP²Ex, including the vertical profiles of NPF frequency in different airmasses. The NPF in background air, and the impact of biomass burning and urban emissions on NPF in tropical marine FT are presented and interpreted in Section 4.

We have made the height ranges more precise. Below 3 km, only a few short periods with elevated $N_{>3\text{ nm}}/N_{>10\text{ nm}}$ were observed within the BL about 50 kilometers downwind west of metro Manila during RF18, and are closely associated with shipping and/or urban emissions. The elevated $N_{>3\text{ nm}}/N_{>10\text{ nm}}$ likely occurred immediately following the dilution of vehicle and engine emissions (e.g., Uhrner et al., 2011; Wehner et al., 2009), and they are not considered as NPF events and therefore excluded from further analyses. This is now clarified in the text.

The summary needs to be improved and extended to conclusion. I asked in the first review for some more interpretation and did not see significant improvement here. Please add conclusions and interpretation. Good, that the dynamic/turbulent aspect was added, but dynamics does not only influence the nucleation rate (which cannot be studied with these data), but furthermore also the growth rate. This can be also a relevant factor. Is there any evidence for the dynamic impact?

Following the referee's suggestion, we have improved the summary and extended it to conclusion. The section is now renamed as "Summary and conclusions" (please also see our response to editor's 3rd comment). We agree that local mixing processes can influence growth rate as well. Unfortunately, we don't have suitable data to examine the particle growth rate, because the flights during CAMP²Ex were not designed to track the particle evolution in the same air parcel (i.e., Lagrangian sampling). The last section now reads:

5 Summary and conclusions

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conditions and altitudes. Most earlier studies found that the NPF typically occurs under the conditions of strong solar radiation around noontime. Here we show NPF can occur in the FT with very low CS in the early morning, despite low actinic flux. Depending on their age, urban emissions can either enhance or inhibit NPF in tropical marine FT. Biomass burning plumes strongly enhance NPF in the outflow region of convective clouds once existing particles are efficiently removed by wet scavenging. Due to the lack of measurements of precursors, the nucleation pathways of NPF in different air mass types remain unclear and should be examined in future studies. The impact of urban and biomass burning emissions on NPF, and the subsequent formation of CCN will also need to be examined in the future by combining field observations and model simulations.

The study should be thoroughly revised in terms of language. Many sentences are too long, some are worded incomprehensibly.

We thank the referee for the comment. We have gone through the manuscript carefully and improved the language.