Response to RC2:

We thank the reviewers for their thoughtful and constructive comments. Please find below the response to each comment or question, including notations of improvements to the manuscript. Reviewer comments are in blue fonts and responses are in black. Changes to the manuscript are highlighted in italics.

- There is a really basic question for me, rising up in the description of instruments. From line 115 on it is explained that two particle counters are measuring number concentrations above 3 nm and 10 nm. In the following table 1, all instruments are listed and there is an addition for the second CPC, that it measures downstream of a thermodenuder and thus, measures the number concentration of non-volatile particles above 10 nm. In my view, this is a significant difference, if N10 is the total number concentration > 10 nm or the number concentration of non- volatile particles > 10 nm. This difference is crucial for all conclusions coming from this paper and thus, it is difficult to formulate a review without knowing what is investigated here. I simply assume, that the thermodenuder is not in use, i.e., not heated, otherwise the data don't make sense to me. If this is not the case and the thermodenuder was heated the interpretation has to be rewritten because it is a different parameter. I went through the individual section in more detail below.

Thanks for catching this. There were two TSI 3772 CPC deployed during CAMP^{2}Ex, one provided the concentration of total number concentration of particles with diameters greater than 10 nm ($N_{>10\ \mathrm{nm}}$), and the other sampled downstream of a thermodenuder to provide the concentration of non-volatile particles larger than 10 nm. We thus added a line in Table 1 describing the measurements of $N_{>10\ \mathrm{nm}}$ by TSI 3772.

Parameter/Variable	Instruments/Methods	Sampling frequency
Number concentration of particles (> 10 nm)	Condensation particle counter (CPC, TSI-3772)	1 Hz

Section 3 Overall Statistical Analysis

- This section should be structured in a clearer way. I do not think that not all subsections fit under 'statistical analysis', e.g., the dependence of Monsoon transition. Here, another headline would help. My question is: are all subsections really needed? What are the main conclusions? The figures are not easy to understand, since every figure is slightly different.

In this section, we present an overview of the NPF events associated with different air mass types as well as the results of k-means clustering analysis, which lay a foundation for the subsequent analyses described in the next section (Section 4). The main conclusions are the increasing trend of NPF frequency with altitude and the variation of the frequency in different air masses. Following the reviewer's suggestions, we have made the following changes to this section:

1. Changed the headline to "Overview of NPF events during CAMP^{2}Ex".

2. Incorporated section 3.3 into section 3.2 and moved Fig. 1 and Fig. 3 to SI; modified Fig. S1 to show the flight tracks and locations of NPF events associated with various air mass types. Fig. S1 is now Fig. 1 in the main text.
3. Modified the figures using condensation sink instead of surface area.

Please refer to the revised manuscript for more details of the revision.

- There are some sentences, which are difficult to digest, e.g. (l. 201f.), 'Most NPF events were observed above 3 km when RH exceeded 50%, and only about 2% of total NPF was observed at 3.5-5 km.' From this, I would conclude that most NPF events occur above 5 km?! Such sentences should be homogenized.

We have edited this paragraph as suggested and it now reads:

There was a total of 19 research flights (RFs) during CAMP²Ex. Figure 1 shows an overview of the flight tracks and the locations where NPF in three major air mass types was observed. These RFs covered the ocean east and west of Luzon Island, and two of them (RF8 and RF18) sampled over Luzon Island and upwind/downwind of Metro Manila. The date and sampling areas of all RFs, together with the duration and key variables of observed NPF events, are presented in Table S1. <u>Most NPF events were observed above 5 km when RH exceeded 50%.</u> A few short periods with elevated $N_{>3\ nm}/N_{>10\ nm}$ (not counted as NPF events) were observed within the boundary layer about 50 kilometers downwind west of metro Manila during RF18, which are closely associated with shipping and/or urban emissions. These NPF events 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 included in further analyses. NPF frequency, defined as the ratio of the sampling time when new particles were observed to the total flight time, decreased drastically starting from RF11 on 19 September and no events were observed from RF12 through RF17 as shown in Fig. S1. This sudden decrease in NPF frequency coincided with the early monsoon transition starting on 20 September (Hilario et al., 2021).

- Figure 2a) it is hard to distinguish low NPF frequencies from zero. At the lowest point, it looks like the frequency in background air is significantly above zero, but it is stated above: 'Figure 2a shows that below 5.5 km, no NPF events were observed in background or BB-influenced air masses (l. 224)' This does not fit for me; such results have to be clear and comprehensible. Please check this paragraph.

Right, the NPF frequency in background air at 3.5 km is above zero. The statement has been revised in the text:

Figure 2a shows that below 5.5 km, NPF frequency is very low (below 3%) and NPF was mostly observed in the urban-influenced air masses. No NPF events were observed in BB-influenced and only minor events took place in the background air masses at ~3.5 km.

- Does Figure 2 include all data?

Fig. 2 includes all data sampled above 3 km as there were no NPF events observed below 3 km.

- Figure 3 includes the same data, but just divided into the two periods? There are too many similar looking pictures, which are not so easy to interpret.

Following the reviewer's suggestion, we have moved this figure to SI and incorporated the discussion into section 3.2 to make it concise to read.

Section 4: Characteristics of NPF in Different Air Mass Types

- In subsection 4.1 NPF events in Background air were analyzed. Here the two different clusters were shown in the following. In the subsection about biomass burning those events from a certain height region were compared with background cases. In the third subsection, the urban NPF cases were compared with non-NPF events. This is really confusing. Here, a more homogeneous way should be selected and maybe one figure containing all three airmasses, maybe in comparison with non-NPF cases can replace some of the figures.

We thank the reviewer for this comment. We tried to use a homogeneous way to compare NPF and non-NPF periods for all three air mass types but find it very challenging. This is because NPF is affected by several interplaying factors. Examining the impact of one factor on NPF often requires comparing measurements with other parameters at the same or similar level, which is often not possible given the data collected during $CAMP^2Ex$. For example, in subsection 4.2 (i.e., BB-influenced NPF), if we directly compare the measurements between NPF and non-NPF periods in BB-influenced air masses, it would be difficult to differentiate the impact of precursors (with CO as proxy) because measurements during non-NPF periods always have higher condensation sink than NPF events do. By comparing between BB-influenced NPF and background NPF, we can more clearly show the role that precursors play in enhancing NPF, given the same level of CS and UV irradiance. This is why we adopted different types of comparisons for different air mass types.

- There are definitely too many and too different figures in the manuscript and this should be changed. Also, the mix of statistical analysis, cases studies as time series and profiles is confusing and should be better structured/motivated.

We have moved a number of figures to SI, including Fig. 1, Fig. 3, Fig. 6, Fig. 9, Fig. 11 and Fig. 12. For the discussion of NPF in urban influenced air masses, time series figures (Fig. 8 and Fig. 10) are kept in the main text, whereas Figure 9 (statistical analysis) has been moved to SI to make the manuscript more concise. Please refer to the revised manuscript for the adjustments.

Summary:

- This section summarizes the most important finding of the study. However, I miss a bit more interpretation. Furthermore, it should be estimated how relevant such NPF processes are at all. Here it should be stated, which type of NPF plays a significant role and which is more a minor process and so on. In general, how relevant is the free tropospheric NPF for this region, for other regions? Is it possible to conclude such a statement from these results?

We thank the reviewer for this suggestion. We used surrogates (i.e., RH, CO) for precursors from different sources and examined the impact of airmass on NPF. However, because there are no measurements of the nucleating species and their precursors (e.g., DMS, NH_3), we are not able to quantify the importance of different nucleation mechanisms. Understanding the role of different nucleation pathways in the FT is clearly very important, but it will require new measurements in future studies.

- In the summary the term condensation sink is used while in figures always surface area is shown. Of course, these parameters are similar, but not identical. I would prefer if CS would be used throughout the whole paper.

Condensation sink based on particle size distribution after hygroscopic growth has been calculated and used as one of the four parameters for the k-means classification. Detailed description of the method is added to section 2.1 (shown below), and discussions can be found throughout the manuscript.

Condensation sink (CS) reflects how quickly condensable vapors will condense on the existing aerosol (Dal Maso et al., 2002). We calculated CS from the ambient aerosol size distribution (Kulmala et al., 2012), which was derived by combining dry particle size distribution measured by FIMS (10-600 nm) and LAS (600-1000 nm), ambient RH, and an average hygroscopicity parameter (κ). Aerosol mass spectrometer (AMS) measurements show that on average, $(NH_4)_2SO_4$ represents 90% of the PM_1 mass above 5 km, where the vast majority of the NPF events was identified. A κ value of 0.53 was therefore applied to calculate particle hygroscopic growth factor at ambient RH (Petters and Kreidenweis, 2007) for each size bin of the combined dry size distribution.

- Maybe some more comparison of the different air mass types is possible, one main difference is probably the condensation sink, but could the authors speculate also about differences in precursor gases?

We have added a figure (Fig. S2) to SI showing the vertical profiles of CS for three major air mass types and a few sentences to section 3.2 to briefly discuss the difference:

Figure S2 shows the vertical profiles of CS for three air mass types. Background air masses have lowest CS on average below 4 km and above 6 km (except for 7-7.5 km) among all three air mass types, whereas BB-influenced air masses have the highest CS at lower altitudes (i.e., < 4 km) and urban-influenced air masses dominates the higher altitudes (> 6 km). The condensation sinks of three air mass types are comparable in between.

There are no measurements of precursor gases during $CAMP^2Ex$. We included discussions of possible precursors involved in different types of NPF events and relevant references (e.g., page 16, line 400-405 for BB-influenced NPF; page 23, line 502 for urban-influenced NPF).

- What about dynamics? Local mixing processes can foster nucleation and growth, is this relevant here?

Very good point. Previous studies have shown new particle formation promoted by mixing in boundary layer, lower troposphere, and tropopause regions, where vertical mixing of air parcels with different temperatures and precursor concentrations can occur. We speculate that mixing may occur in the outflow regions where the air parcels from lower altitudes are pumped aloft and mixed with the surrounding air. Such mixing could promote NPF observed during $CAMP^2Ex$. In this study, the NPF was identified using elevated $N_{>3nm}/N_{>10nm}$. As the observation of particles between 3 nm to 10 nm typically occurs several hours following the initial particle formation, we are not able to pinpoint the exact locations of initial particle formation and isolate the impact of the mixing processes on NPF.

We have added the following sentences to the 2nd paragraph in section 3.2 to briefly discuss the potential roles of mixing processes:

This suggests NPF in outflow regions and detrainment layers of convective clouds, which is confirmed by the flight video, is also consistent with earlier studies (Clarke et al., 1998; Perry and Hobbs, 1994). <u>Previous studies show that the mixing of air mass with different temperature and precursor concentrations can lead to enhanced nucleation rates (Khosrawi and Konopka, 2003; Nilsson and Kulmala, 1998; Nilsson et al., 2001; Wehner et al., 2010). In the outflow regions, the mixing of cloud outflow and surround air may also contribute to the observed NPF events.</u>

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