Response to RC1

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.

- One general concern, which needs to be at least thoroughly discussed in the manuscript, is that the authors often connect the observation of sub-10 nm particles (elevated $N_{>3nm}/N_{>10nm}$ ratio) directly to the observed external conditions and link this occurrence of "NPF" to them. However, in the FT new particle growth rates might be just in the order of <1 nm h^{-1}, which means that the observation of 3-10 nm particles could originate from particle formation processes which already occur over several hours (up 15-20 hours, if we assume GR=0.5 nm h^{-1} and the growing nucleation mode to be located at around 8-10 nm, which is not resolved by the simple $N_{>3nm}/N_{>10nm}$ ratio due to missing size-distribution information). In that case, the current conditions under which the sub-10 nm particles are observed might not at all representative for the conditions under which the new particles might have been forming. The authors should discuss potential implications of this within their analysis.

Response: This is a very good point. Ideally, the concentration of incipient particles (i.e., particles with diameter around 1.5 nm) should be used to identify new particle formation (NPF) events. However, given the challenges of measuring particle concentration below 2 nm onboard research aircraft, many airborne studies have used the ratio of $N_{>3nm}/N_{>10nm}$ (e.g., Zheng et al., 2021) and/or $N_{3\text{-}10\,nm}$ (e.g., Crumeyrolle et al., 2010) to characterize NPF events.

As the reviewer pointed out, the observation of 3-10 nm particles likely occurs several hours after the particle formation. As incipient particles are efficiently removed by coagulation inside clouds, we expect the air mass with elevated $N_{>3nm}/N_{>10nm}$ remained cloud free and did not experience precipitation since the recent particle formation. Therefore, condensation sink (CS) and relative humidity (RH), which are among the NPF related parameters examined in this study, are unlikely to vary drastically over a period of several hours following the particle formation. New particle formation and subsequent particle growth can lead to an increase of CS. For elevated $N_{>3nm}/N_{>10nm}$ observed under conditions of low CS, the formation of new particles likely had occurred with comparable or even lower CS. UV irradiance has a strong diurnal variation and depends on the cloud condition, and it can change substantially over a period of several hours. In this study, most NPF events (i.e., elevated $N_{>3nm}/N_{>10nm}$) were observed at noontime under higher levels of UV irradiance compared to the non-NPF periods at the same altitude, consistent with earlier studies showing that solar radiation was generally higher during NPF event days compared with non-event days. Some of NPF events were observed under conditions of low UV irradiance, and the potential mechanisms are discussed in Section 4.1.

Following the suggestions, we have included the discussion below in section 3.3:

Because it takes some time for incipient particles to grow into the 3-10 nm size range, the NPF events identified here using $N_{>3nm}/N_{>10nm}$ *value are likely several hours after the formation of the new incipient particles, depending on the actual growth rate. As the incipient particles are* *efficiently removed by coagulation inside clouds, we expect that air masses with elevated $N_{>3nm}/N_{>10nm}$ remained cloud free and did not experience precipitation since the recent particle formation. Therefore, CS and RH, which are among the NPF related parameters examined in this study, are unlikely to vary drastically over a period of several hours following the particle formation. New particle formation and subsequent particle growth can lead to an increase of CS. For elevated $N_{>3nm}/N_{>10nm}$ observed under conditions of low CS, the formation of new particles likely have occurred with comparable or even lower CS. UV irradiance has a strong diurnal variation and depends on the cloud condition, and it can change substantially over a period of several hours. In this study, most NPF events (i.e., elevated $N_{>3nm}/N_{>10nm}$) were observed at noontime under higher levels of UV irradiance compared to the non-NPF periods at the same altitude, consistent with earlier studies (Kerminen et al., 2018) showing that solar radiation was generally higher than during NPF event days compared with non-event days. Some of NPF events were observed under conditions of low UV irradiance, and the potential mechanisms are discussed in Section 4.1.*

- As this study focusses on NPF it would be very useful to use the quantity of condensation sink (CS in s^{-1}, as defined by Dal Maso et al. 2015) instead of surface area. If the CS is even calculated using assumptions on the hygroscopicity of FT particles, it would much better represent the actual sink of condensable vapors and, related to it, the coagulation sink of newly formed clusters. This would incorporate the RH information into that parameter (high RH means also even higher sink typically at the same dry aerosol surface area. Related to that: Do FIMS and LAS actually dry the sample? I am missing that information from the Methods section). This would significantly help to relate the observations of sub-10 nm particles in the FT to NPF at many other locations where CS is reported. Related to comment 1) a comparison of CS to a potential GR then also directly gives an approximation of the survival probability.

Response: We thank the reviewer for this suggestion. The FIMS and LAS measured dry aerosol size distribution. We derived an average hygroscopicity parameter (κ) using AMS measurements for aerosols sampled above 5 km, where the vast majority of the NPF events was identified. The CS was derived from the ambient aerosol size distribution, which was calculated from the average aerosol hygroscopicity, ambient RH, and measured dry aerosol size distribution. The aerosol surface area was replaced by CS in all relevant figures and discussions were modified accordingly throughout the main text. We reworked the line 133-136 as follows to describe the approach of calculating CS:

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.

We also included the comparison of CS to previously reported values in section 3.2, where Fig. 2 presents the vertical profile of CS for NPF and non-NPF periods of the whole mission:

Figure 2 shows that most NPF events occur when CS is below 0.002 s^{-1}. For NPF events observed above 6.5 km, the median CS value is mostly below ~ 0.001 s^{-1}, comparable to the CS below 8 × 10^{-4} s^{-1} globally in the tropical mid-FT reported by Williamson et al. (2019).

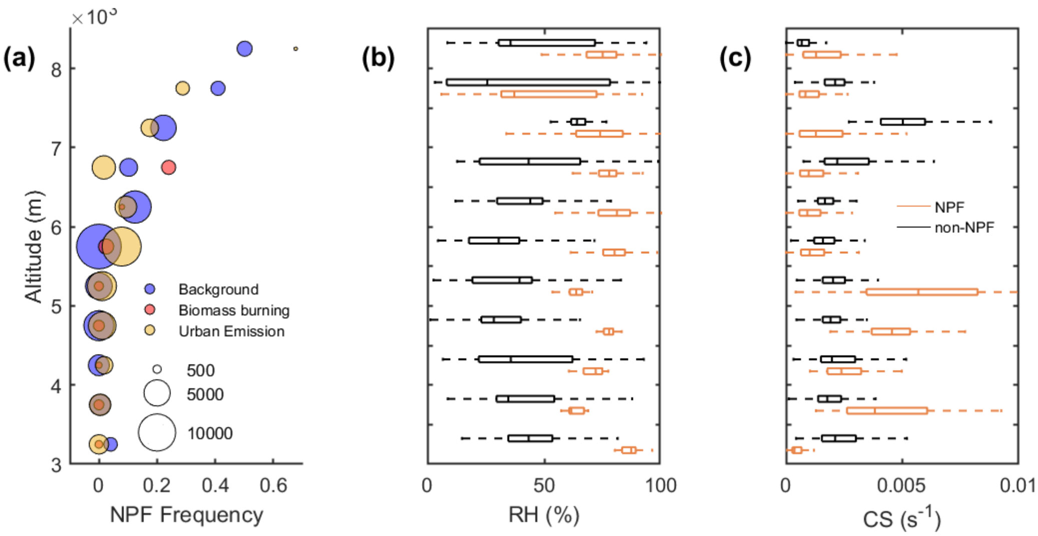

Figure 2. *(a) The vertical profile of NPF frequency for the three air mass types. NPF Frequency is defined as the ratio of total duration of NPF period to the total sampling time outside of the clouds for each airmass type. Also shown are the comparison of (b) relative humidity (RH) and (c) condensation sink (CS) between NPF and non-NPF periods, where black denotes non-NPF and orange denotes NPF.*

As the reviewer points out, the survival probability depends on both growth rate and coagulation sink (Kulmala et al., 2012; Kerminen et al., 2018; Kuang et al., 2009; Westervelt et al., 2013). However, we cannot derive the particle growth rate from the measured aerosol size distribution because the CAMP^{2}Ex flights were not designed to track the aerosol evolution in the same air masses (i.e., Lagrangian sampling).

- I am missing the clarity in the analysis, especially from Section 4 onwards, where I find many Figures difficult to read (and potentially unnecessary for the main text), while other Figures would have been helpful. I have some major points here:
  My biggest scientific concern related to that is that the authors often compare conditions of NPF of a certain cluster and/or attributed air mass origin with conditions of no-NPF at the same altitude. However, as altitude is not the only variable which leads to the classification of a certain NPF event in a certain cluster and/or attributed air mass origin, this comparison is in my opinion not very useful. I thus suggest leaving the no-NPF periods from Fig. 4b-f and delete Fig. 9 (I think Fig. 8 already shows the most important conclusion for Section 4.3.1).

Figure 4 (now as Fig. 3) has been modified as suggested (see modified Figure below). All the non-NPF data have been removed and panel c now shows CS.

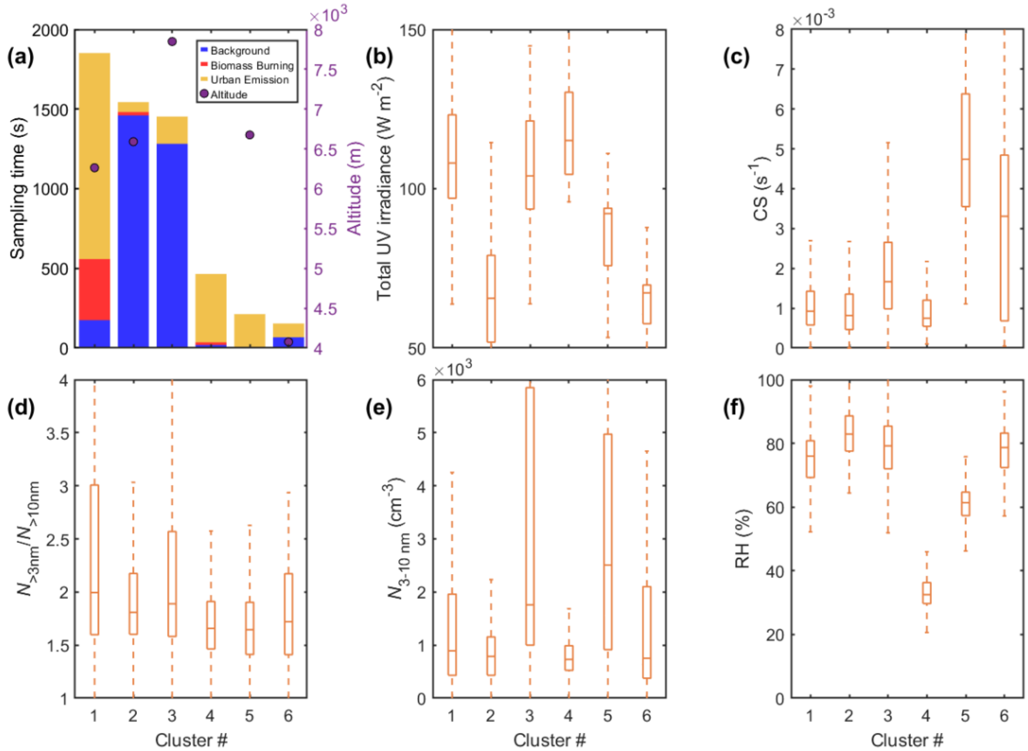

Figure 3. (a) Amount of data classified into each cluster and contributions of different air mass types. The other five panels compare the clusters in terms of (b) total UV irradiance, (c) CS, (d) the ratio of number concentration of particle larger than 3 nm to that of particle larger than 10 nm ($N_{>3\ nm}/N_{>10\ nm}$), (e) number concentration of particles between 3 and 10 nm ($N_{3\text{-}10\ nm}$) and (f) RH.

Fig. 9 supports the conclusion of Section 4.3.1 from a statistical perspective. We have moved Fig. 9 to SI.

- 6 and Fig. 11, Fig. 12 are in my opinion supportive material but no major results and could easily be moved to the SI. I also consider Fig. 1 not as the most important Figure to start the manuscript with. It remains unclear what is meant by "sampling data count". Why not color the bar plot with the amount of background, biomass-burning and urban emission related NPF and no-NPF observed during the flight and add it as an additional panel to Figure 2.

Following the reviewer's suggestion, we have moved Figures 6, 11 and 12 to the SI. Regarding Fig. 1, sampling data count refers to the total amount of 1-second data collected during each flight. Figure 1 has been revised accordingly. However, including Fig. 1 as an additional panel of Fig. 2 will make Fig. 2 very crowded. Therefore, Figure 1 is now moved to SI instead. The revised Fig. S1 is shown below, with air mass types colored the same way as in Fig. 3.

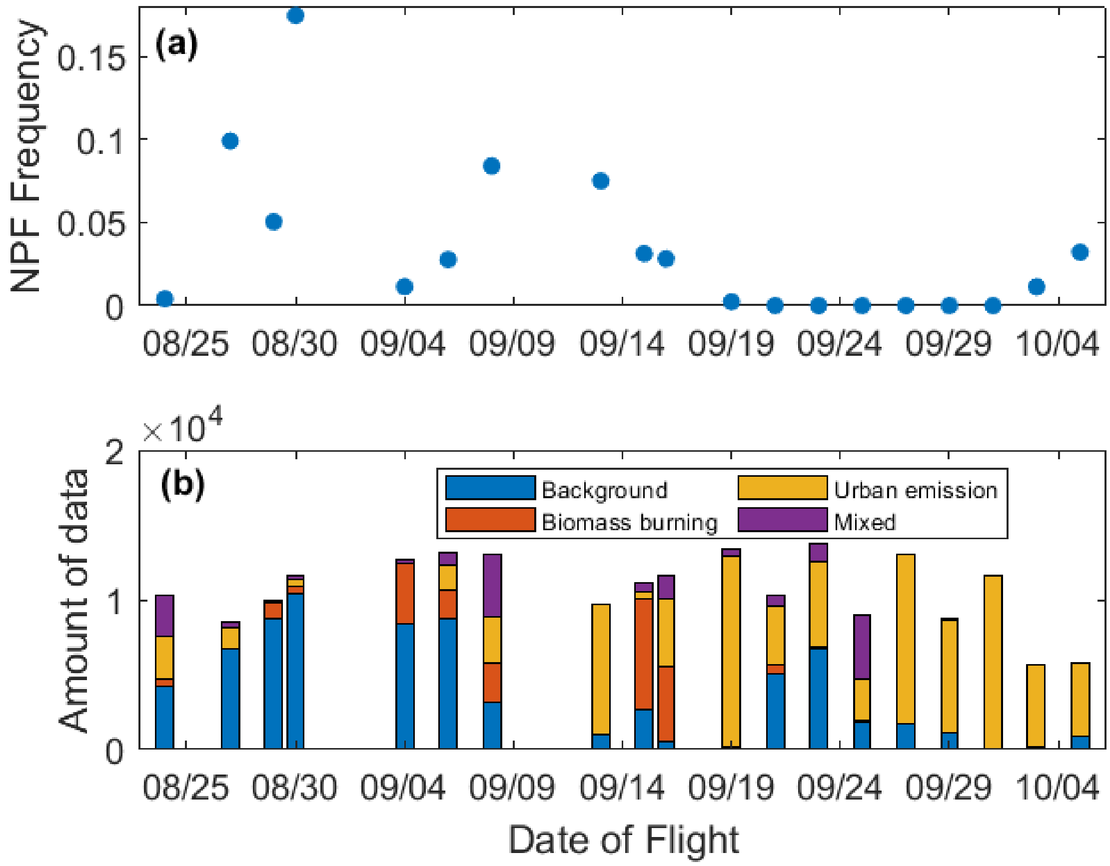

Figure S1. (a) NPF frequency for each flight, i.e., the ratio of NPF events to the total amount of data for each RF. (b) Bar chart of the amount of data sampled during each RF and color coded by air mass types.

- To put all the flights and observations into a clearer relation I suggest making an additional version of Figure S1 (maybe a second panel), where all flights are represented as black lines when no NPF is observed and colored when NPF is observed with the same coloring as in Fig. 4a (blue for background, yellow for urban and red for biomass-burning). Moreover, please add a legend to the already existing panel relating the current colors to the number of the RF. Please also improve the resolution of that Figure significantly to make it better readable. If all the updates are made according to my suggestions, Fig. S1 could even move to the main MS as it gives the reader a quick geographical overview. Please also indicate the position of Manila on the map!

This is an excellent suggestion. Figure S1 has been modified as suggested and moved to the main text as Fig. 1 (shown below).

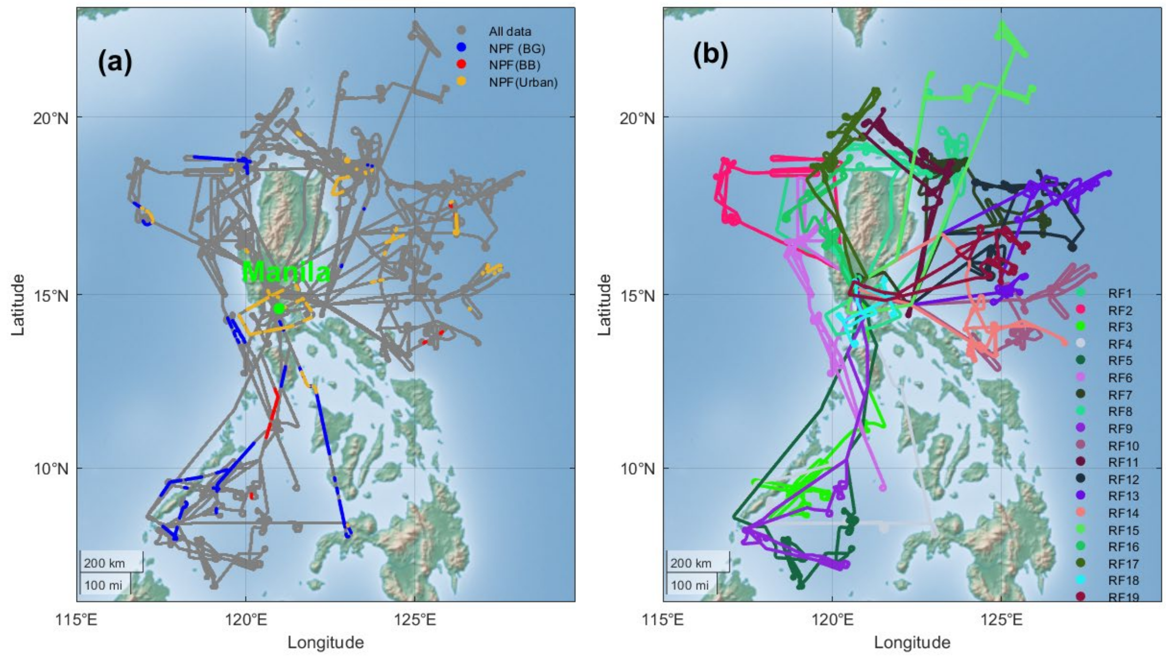

Figure 1. *(a) NPF events during the whole mission color coded by three air mass types (background, BB-influenced and urban-influenced). (b) Locations of 19 flight tracks.*

Section 3.1 now reads:

There was a total of 19 research flights (RFs) during CAMP²Ex. <u>Figure 1 shows an overview of the flight tracks and the locations where NPF in three major air mass types were observed</u>. 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. Most NPF events were observed above 5 km when RH exceeded 50%. A few short periods with elevated $N_{>3nm}/N_{>10nm}$ (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).

Minor comments

- Page 2, 56-57: Please provide a reference for that statement.

A reference has been added in line 56-57 and the text now reads:

Essentially all long-term surface measurements show that the average solar radiation intensity is stronger during NPF event days compared with non-event days (Kerminen et al., 2018).

- Page 3, line 90: "from the perspective of galactic cosmic rays". What is this supposed to mean? GCRs are known to enhance NPF of weakly binding systems such as H_2SO_4+NH_3 and HOMs. The work of the CLOUD team (Kirkby et al., 2011 and 2016, Nature) should be mentioned as they have provided the most thorough investigations of the role of GCRs in NPF so far.

Thanks for suggesting the reference. Line 89-91 now reads:

Kirkby et al. (2011) found that ion-induced binary nucleation associated with galactic cosmic ray can occur in mid-troposphere but is negligible in the boundary layer, while the strongest aerosol formation takes place in upper troposphere over tropic ocean (Kazil et al., 2006).

- Page 3, line 107: In accordance with the guidelines of ACP, please refrain from citing unpublished references. See guide for authors: "Works cited in a published manuscript should be published already, accepted for publication, or available as a preprint with a DOI".

The paper has been published in BAMS online and reference is now added: *Reid, J. S., and Coauthors, 2023: The coupling between tropical meteorology, aerosol lifecycle, convection, and radiation, during the Cloud, Aerosol and Monsoon Processes Philippines Experiment (CAMP2Ex). Bull. Amer. Meteor. Soc., https://doi.org/10.1175/BAMS-D-21-0285.1, in press.*

- Page 5, line 134-135: "combined size distribution from multiple instruments, including FIMS and LAS". Are FIMS and LAS the only instruments combined here than delete "multiple instruments, including", or were there other instruments incorporated in that combined size distribution than mention them explicitly before. Were the size-distributions just added as the instruments covered different size-ranges or was there some combining instrument inversion applied?

We combined size distributions from FIMS (10-600 nm) and LAS (600-1000 nm). No combined data inversion was applied. We have clarified this in the revised manuscript. Please refer to the response to the 2nd major comment regarding the calculation of CS.

- Page 5, line 149: How is the uncertainty of the ratio defined? By the variance of the data within the 10 second interval, or by some pre-set error on the estimates of $N_{>3nm}$ and $N_{>10nm}$? Please specify.

The uncertainty of the concentration ratio was derived from uncertainties in $N_{>3nm}$ and $N_{>10nm}$ using uncertainty propagation. The uncertainties in $N_{>3nm}$ and $N_{>10nm}$ are calculated based on the counting statistics of the two CPCs. The approach is detailed in Zheng et al. (2021) as referenced in line 143-144.

- Page 6, line 160-161: Such periods are often called "undefined" in typical NPF studies.

We agree. On the other hand, the analysis is focused on identified NPF events in this study. We prefer not to introduce additional definitions that are rarely used in the manuscript.

- Table 2: It could be useful to also give the data ranges for the different mean values and clusters.

Standard deviation has been added to Table 2 (see below for revised Table 2).

Table 2. General statistics of key parameters for the 6 clusters identified using k-means classification.

Cluster #	Number of events	Amount of 1-s data	Mean±std altitude, m	Mean±std temperature, ºC	Mean±std UV irradiance, W m^{-2}	Mean±std RH, %	Mean±std CS, 10^{-3} s^{-1}
1	35	5550	6104.6±591.9	-4.6±3.3	108.8±13.6	75.4±9.0	1.1±0.5
2	20	3870	7708.8±433.2	-15.2±2.4	104.5±13.1	79.8±8.5	2.0±0.7
3	13	3960	6392.4±369.8	-7.3±1.8	60.9±14.4	82.6±7.0	1.1±0.5
4	9	1190	7532.1±438.2	-12.9±2.5	118.8±21.2	33.3±13.5	1.2±0.6
5	11	790	6698.5±650.7	-10.3±4.2	93.7±23.6	61.3±6.3	5.1±1.2
6	7	400	3959.3±671.3	4.2±4.3	58.1±24.1	74.2±10.5	2.9±2.0

- Page 7, line 183-189: In the absence of an accessible version of DiGangi et al. (see above comment) this needs to be more detailed as this is a central part of how the different NPF occurrences have been specified.

Joshua DiGangi suggested that the description of the emission flag data can be cited here https://doi.org/10.5067/Airborne/CAMP2Ex_TraceGas_AircraftInSitu_P3_Data_1.
We reworked line 183-184 and it now reads:

To investigate the impact of air mass type on NPF, we classified air masses sampled during CAMP2Ex into three types (details can be found at https://doi.org/10.5067/Airborne/CAMP2Ex_TraceGas_AircraftInSitu_P3_Data_1.).

- Page 7, line 186: Here you speak about 4 regimes, but mixed urban/biomass burning is not mentioned again at any later stage in the manuscript and does not appear in Fig. 4.

Yes, the 4 regimes were defined by the reference cited above, but we simply focused on the 3 main regimes (i.e., background, biomass burning and urban emission) in the analysis. We clarified this in the revised line 193-194:

In this study, we use the first three regimes to investigate the impact of air masses on NPF by focusing on NPF events observed in background, biomass burning and urban-influenced air masses.

- Page 8, line 221-223: If we call these events undefined, this could be used here and would make the sentence better accessible to the reader.

We agree the sentence is a little bit long. On the other hand, we feel it is better to clearly spell out what data are used to calculate the statistics for non-NPF periods above 7.5 km here.

- Page 9, line 233: "impact" might not be the best word to use here as surface area and airmass as NPF is a result of airmass, surface area and other factors. Maybe "interplay"?

We have changed "impact" to "relationship". The sentence now reads:

The altitude dependence of the relationships among air masses, CS, and NPF implies the competing influences from different processes (i.e., production and removal of nucleating species) that vary with altitude, which will be further discussed in Sect. 4.

- Figure 2: As you discuss the panels in the different order in the text, it makes sense to swap the current panels b and c.

Figure 2 has been modified as suggested.

- Page 10, line 271: "was elevated compared to all the other clusters" is also true and maybe even more important.

Good point. Line 270-271 now reads:

The NPF events classified as cluster #5 have the highest CS compared to the other clusters and were mostly observed during RF18 and RF19.

- Page 10, line 272: These were the MT flights. Could be mentioned that those were at overall different meteorological conditions.

We edited this sentence to relate cluster #5 to the different meteorological conditions resulting from monsoon transition:

The NPF events classified as cluster #5 have the highest CS compared to the other clusters and were mostly observed during RF18 and RF19. Both flights took place near the end of CAMP^{2}Ex during monsoon transition, when airmass origins and meteorological conditions are likely different from those of earlier flights.

- Page 11, line 275: should be 1, 3 and 4!

This has been corrected and now reads:

Figure 3b shows that most of NPF occurred with high actinic flux (indicated indirectly by the UV irradiance data during this campaign), as in clusters 1, 3, and 4.

- Page 13, line 329 – Page 14, line 344: This paragraph is difficult to follow and rather lengthy. As I suggest moving Fig. 6 to the SI, it could be shortened: The main message here is: cluster #1 has high UV, occurs at noon and has a higher CS. In contrast, cluster #2 has low UV, is at the morning and has a lower CS. Investigation of the occurrence of times with such low CS shows that such periods occur more often in the early morning than in the late afternoon and the NPF frequency for mornings is higher than for the late afternoons, which indicates that the newly formed particles are indeed formed in the early morning and are not related to NPF from the previous day.

Following the reviewer's suggestions, we have moved Fig. 6 and detailed discussion to SI. This paragraph (line 329-line 349) has been shortened and now reads:

One possible explanation is that these new particles were formed during the previous daytime under high UV irradiance/actinic flux, survived scavenging overnight and were detected the next morning. However, the low CS conditions are much more prevalent in the early morning than in the late afternoon (Fig. S5 and related discussion). In addition, the frequency of NPF in the early morning is about 20 times higher than that in the afternoon, suggesting that new particles observed most likely formed in the morning instead of the day before. The NPF in the early morning is likely made possible by the much lower CS despite the lower UV irradiance and actinic flux. We speculate the prevalence of low surface area in the early morning is due to a combination of wet scavenging and less convection overnight.

- Page 16, line 398-399, Fig. S4: I do not see these trends from Fig. S4. N>100 nm seems to be quite similar between the two periods. What do you mean by "as high as when NPF was absent". Please clarify. Please also change the x-axis label of the Fig. S4 (strange unit, what does 27 hours mean? Just put a normal time axis there).

The term "the other segment" should be referring to the segment where no NPF was observed. The sentence is revised to further clarify:

For the other segment (Fig. S7, 11:15-11:25), NPF was absent and the concentrations of non-volatile particles and larger particles (> 100 nm) were three times as high as those of the NPF events (Fig. S7, 12:45-12:55).

- Page 17, line 402: Here the focus could be more on NPF by adding "(…) and mixtures of sulfuric acid ammonia and organic vapors are shown to be efficient new particle formation agents (Lehtipalo et al., 2018, Sci. Adv.)" right after the reference to Ahern et al. (2019).

We have modified the text accordingly. It now reads:

It remains unclear which nucleation pathway dominates particle formation observed in BB-influenced air mass, since organic vapors, ammonia (Hegg et al., 1988) and sulfuric acid can directly or indirectly originate from biomass burning plumes and contribute to formation of secondary aerosols (Ahern et al., 2019), <u>and mixtures of sulfuric acid, ammonia and organic vapors have been shown leading to strong NPF (Lehtipalo et al., 2018).</u>

- Page 23, line 503: Please also refer to newer studies investigating NPF in highly polluted environments, e.g., Yao et al., 2018 (Science).

The sentence has been edited as suggested:

The mechanism for this type of NPF is likely similar to those observed in polluted urban boundary layers (Alam et al., 2003; Zhu et al., 2014; Yao et al., 2018), where high concentrations of precursors make NPF possible despite relatively high condensation sink conditions.

- Page 23, line 504-506: Could temperature be decisive here? Especially when organics are involved in the formation process there is an interplay between the degree of oxidation and volatility which both depend strongly on temperature. If oxidation occurs at lower altitudes and high T, highly oxygenated molecules might form which are however still not able to condense onto the smallest clusters at that temperature, but during updraft and cooling of the airmass this might become possible. This should be discussed in the manuscript. You could refer to Stolzenburg et al. 2018 (PNAS) for these competing processes.

We agree that temperature could also play an important role in the nucleation of organic species. The absence of the NPF events at low latitudes (i.e., below 3 km) is likely due to the combination of warm temperature and high condensation sink. A higher temperature leads to increased volatility of secondary organic species, slowing down the growth of the newly formed clusters. Line 504-506 has been edited and now reads:

The absence of NPF below 2 km may result from a combination of higher CS and warmer temperature compared to those in the detrainment layers at higher altitudes. Stolzenburg et al., (2018) show that temperature impacts the growth by organics via competing processes. While a higher temperature leads to faster reaction rate and high concentration of highly oxidized molecules, it also strongly increases the volatility of organic species, therefore slowing down or even inhibiting the condensation of organic vapors onto the incipient clusters.

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