We would like to appreciate the reviewer for providing valuable comments on our manuscript, and we have carefully addressed these comments point-by-point as follows. Please find the response (in red) to each comment below.

Referee comments:

This paper examines the effects of hygroscopicity, surface tension and aerosol processes on the NPF contribution to Nccn, based on field observations and modeling of three NPF events at a rural site in southern China. The study results and implications are of interest to ACP readers. The adopted experimental setup and methodology are well-established, comprehensive, and thus reasonable. The manuscript is generally well-written and organized, though some data presentation, interpretation and discussion can be improved.

Major comments:

1. Because there are only three NPF events, quite thoroughly, analyzed in this study, it is crucial that the authors should somewhat discuss the representativeness of those three NPF events. The dominant mechanisms driving NPF vary with time and location.

Reply: We appreciate the reviewer for this valuable suggestion. We totally agree with the reviewer that the dominant mechanisms driving NPF may vary temporally and spatially. We add a discussion on the representativeness of the three NPF events in lines 566-579 in section 3.4 as follows,

"It should be noted that the three NPF events discussed in this study were generally "Class I" regional NPF events, for which the growth rate and formation rate could be obtained with high confidence (Dal Maso et al., 2005). Other types (i.e., Class II proposed by Dal Maso et al. (2005)) were not considered since their growth rates and formation rates are extremely difficult to be determined, leading to high uncertainties in model simulation of these events. In addition, we did not include the "transport" type of NPF events, for which new particles were formed somewhere else and then transported to the measurement site, because the model ignores the impact of transport. Some events belonging to "Class II" type and "transport" type were observed during the campaign (Fig. S10). For the "Class II" type (Fig. S9 a), the number concentration and diameter of the nucleation and Aitken mode particles vary significantly. For the "transport" type (Fig. S10 b), the concentration of 3-30 nm particles at 10:00-12:00 LT was much lower than that of 30-70 nm at 12:00-20:00 LT, indicating the impact of transport. Investigation on the contribution of other NPF types to the N_{CCN} is needed in future studies. Moreover, this study only analyzed three NPF events as representatives of Class I type in the PRD region, and more field campaigns in other regions and seasons are also needed to identify the major impact factor.

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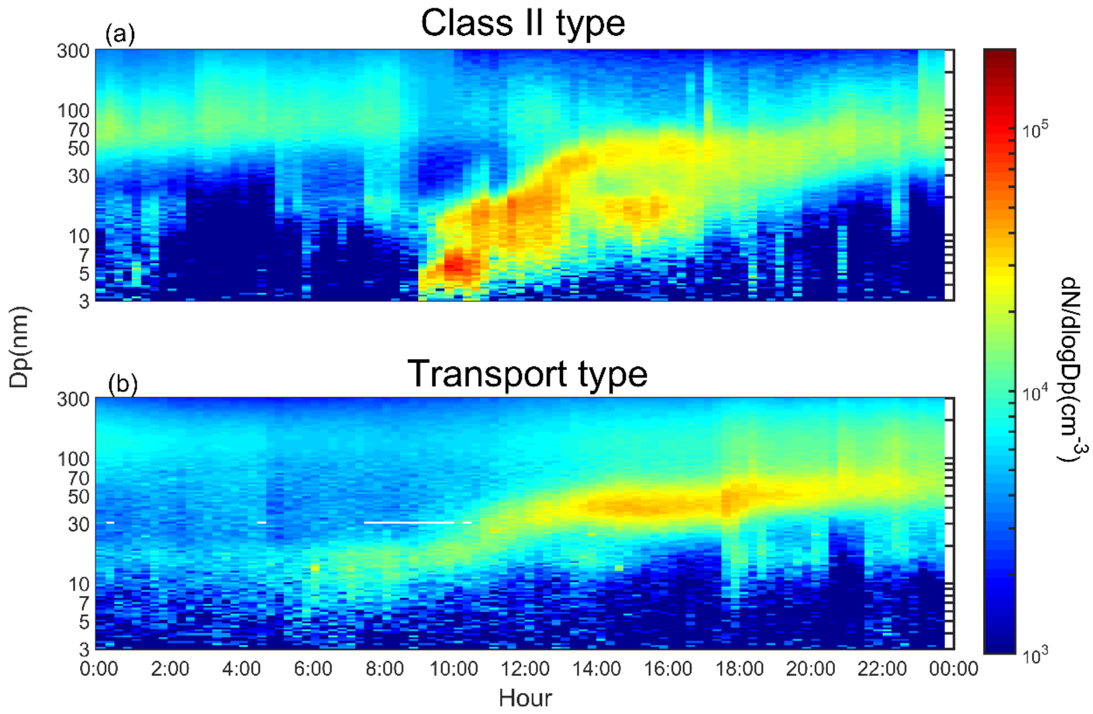

Figure S10. The “Class II” type NPF event (a) and “Transport” type NPF event (b) observed on 9th September and 14th November during the Heshan Campaign, respectively.

2. Although the hygroscopicity and the estimated surface tension were derived under different water saturation ratio (undersaturated vs. supersaturated), two are interlinked with each other. The discussion in Section 3.2 seems to treat the two as unrelated factors. Also, e.g., in the abstract, the authors suggested the surface tension is more important than hygroscopicity (line 37). It is recommended that the authors elaborate/clarify on the rationale of discussion based on adjusting only the surface tension in kccn (but not kHTDMA?), or kHTDMA is the “reference” hygroscopic parameter, and the potential relationships between the two. The discussion and statements should be rephrased to accurately describe the observed cause and effect in a relationship.

Reply: We agreed that the impact of surface tension on the hygroscopicity growth under subsaturated should also be considered. We have recalculated the κ_{CCN} and κ_{HTDMA} by adjusting the surface tension and found that the κ_{HTDMA} was slightly changed with the change of surface tension (Fig. S3). The κ_{HTDMA} $\sigma_{s/a}$* was not changed considerably with $\sigma_{s/a}$*=0.060 N m^{-1} and thus this value ($\sigma_{s/a}$*) was still adopted in the following discussion.

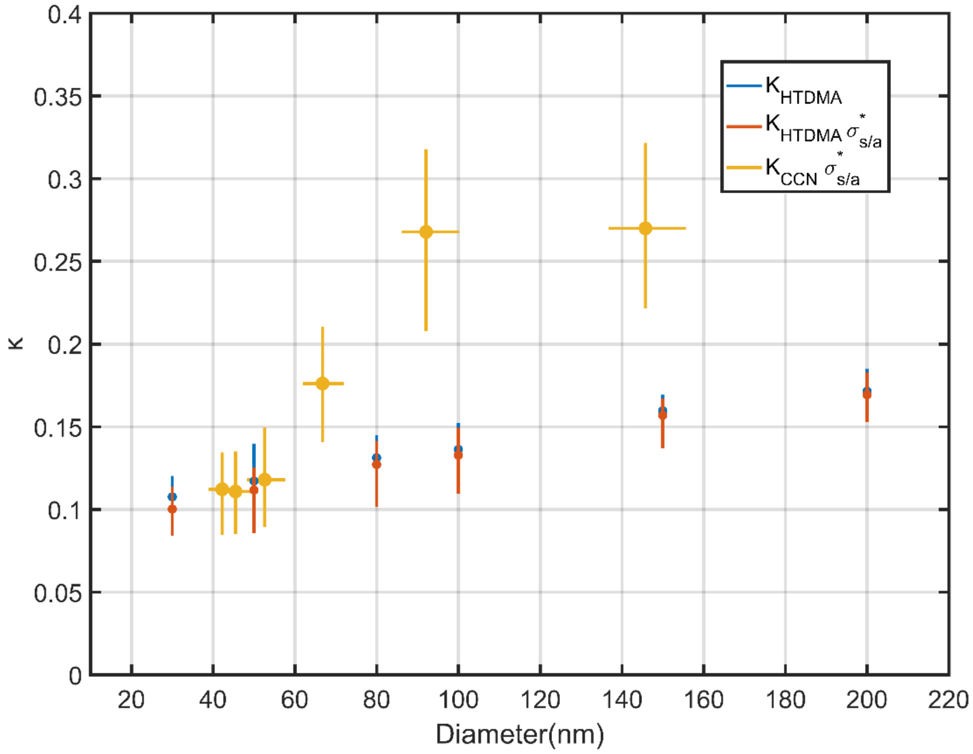

Figure S3. The median and interquartile κ_{HTDMA} and κ_{CCN}. The red and yellow line represent the κ value calculated based on $\sigma_{s/a}$* (0.060 N m^{-1})

We have modified the discussion in section 3.2 in lines 334-338,

"Previous studies showed that surfactants could modify the ability of water uptake, leading to discrepancies of κ values between measurements using techniques under different water saturation conditions, e.g., sub-saturation (HTDMA measurements) or supersaturation (CCNc measurements) (Cai et al., 2018; Wex et al., 2009; Rastak et al., 2017; Ruehl and Wilson, 2014)."

and lines 363-370:

"A surface tension value ($\sigma_{s/a}^*$=0.060 N m^{-1}) was adopted to calculate both the κ_{CCN} (denoted as κ_{CCN} $\sigma_{s/a}^*$) and κ_{HTDMA} (κ_{HTDMA} $\sigma_{s/a}^*$) using Eq. (2) and Eq. (4), respectively. No significant changes of κ values (i.e., from 0.11 to 0.10 for 30 nm particles) were found from TDMA measurements (Fig. S3), while the κ values from CCNc measurements using this surface tension value ($\sigma_{s/a}^*$) were still lower than those using pure water assumption and the differences became larger with increasing particle sizes, implying that the surface tension is dependent on particle diameter. It also implies that the κ value was more susceptible to surfactants under supersaturation condition, which can lower the D_{50} of the particle for facilitating CCN activation."

3. In Section 3.2, lines 389-397, the use of the term "newly-formed" particles should be more specifific and consistent, whether it refers to 40-45 nm particles, or « 30-40 nm particles. It is unclear that the k values discussed therein are kccn or kHTDMA. The "gradual" drop of sulfuric acid (SA) does not necessarily imply it is responsible for the increase of k values because SA condensation is considerably more favorable with larger pre-existing particles, and/or the consideration of oxidant

availability.

Reply:

We thank the reviewer for the suggestion.

(1) The size range of “newly-formed” particles is difficult to define, owing to the continuous growth processes during the event. In order to avoid confusion, these particles were referred to as “newly grown particles”, since they were grown from newly-formed particles. We have modified corresponding sentences in lines 411-414:

“The hygroscopicity of newly-grown particles can have significant impact on the N_{CCN} during the NPF event. During the campaign, the minimum particle size of CCN activity measurement was about 40-45 nm (at 1.0% SS), thus the hygroscopicity of this size range was used to present the property of newly-grown particles, when they grow up to this size range.”, and line 418-423, “It should be pointed out that the high κ values during 10:00~12:00 LT did not represent the hygroscopicity of newly-grown particles which were primarily composed of particles much smaller than 30-40 nm. Those new particles grew to about 40-50 nm at 14:00-16:00 (Fig. 1a and Fig. 3) and their κ values were obviously lower than the average ones, implying that the organic vapors could play an important role during growth of new particle as discussed in Section 3.1.”

and lines 430-431, “As discussed in section 2.3.4, the dynamical processes for new particles during nucleation events are governed by the population balance equation (Eq. (13)).”

(2) The κ discussed in this section was only limited to the κ_{CCN} measured at 1.0% SS because the time resolution of HTDMA measurement was low (about 4 hours). We agreed that condensation of gaseous H_2SO_4 might not be responsible for the increase of κ values, and other organics vapors (e.g., amines) could be a possible reason for the increasing hygroscopicity. Based on the above reasons, we deleted the sentences in line 395-397 “The calculated H_2SO_4 concentration peaked at about 10:00-11:00 and subsequently decreased to a low level (about 0.5×10^{7} cm^{-3}) until 16:00, implying that the increase of hygroscopicity was related to the condensation of H_2SO_4 vapors.”. We have also modified the discussion in lines 414-418:

“In general, the κ_{CCN} values for 40-45 nm particles were significantly higher (corresponding to much higher hygroscopicity) during early event period than during non-event and other event periods (Fig. S4a). Hence, we adopted a minimum size range of 40-45 nm particles for CCN activity measurements (at about 1.0% SS) to represent typically growth of newly-formed particles to this size range during the campaign.”

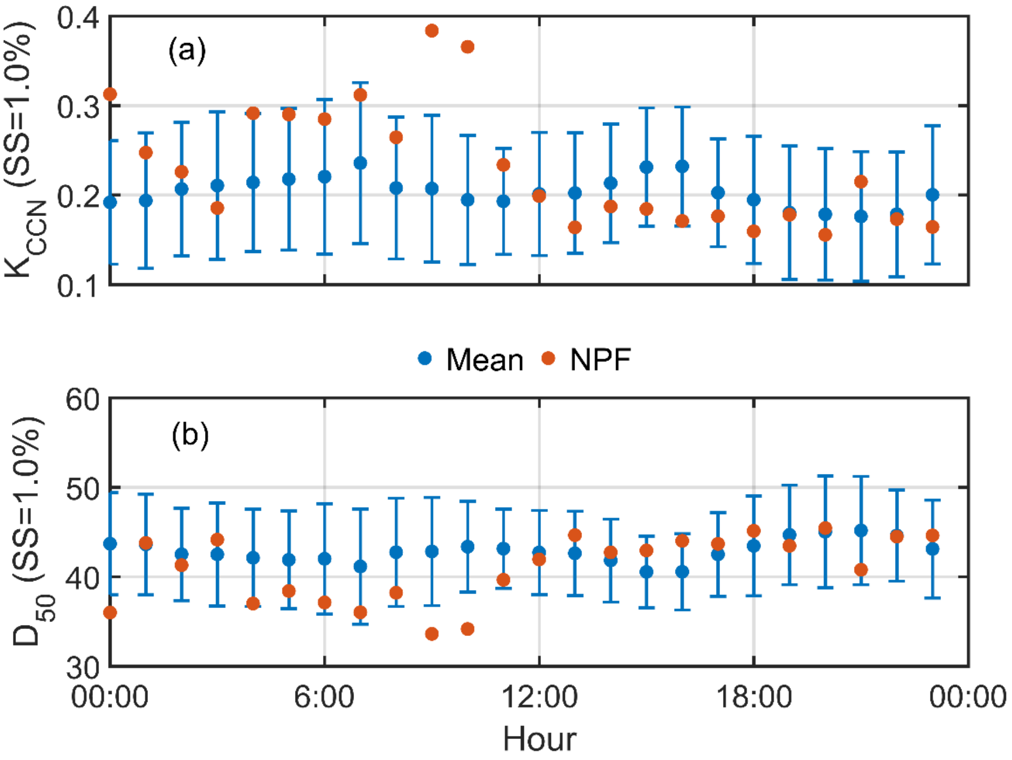

Figure S4. The diurnal variation of κ (a) and D_{50} (b) measured at 1.0% SS. The blue color represents the average value during the campaign. The red color represents the value during the NPF events.

4. Section 3.3 seems to be an add-on modeling analysis of the NPF events not strongly or quantitatively linked to the hygroscopicity and surface tension. The derived conclusions about formation/growth rates and coagulation loss are not new, but expected. This analysis is then extended to Section 3.4 where the three NPF events from two locations are compared. My concerns are (1) the modeled-Nccn deviate notably from measured-Nccn (Figs 5, 6 and 9), and (2) how the findings herein are related to the other subjects of interest regarding the hygroscopicity and surface tension. Please clarify.

Reply:

(1) There are two possible reasons for the deviation between the modeled N_{CCN} and measured N_{CCN}. Firstly, our model does not consider transport and local primary emissions which may partly contribute to the deviation. For example, a significant mode peaking at about 100 nm was observed for the Panyu event, suggesting impact of air mass transport or local emissions. Secondly, we assume constant background particle distribution during NPF events, while actual background PNSD varies substantially from one event to another for the three chosen NPF events. Noticeably, significant variation of Aitken mode was observed for the two Heshan events, leading to failure of reproducing the concentration trend of 10-60 nm particle at the early event stage. To clarify, we have modified the discussion in lines 535-543,

"For the October 18 event, however, the model underpredicted the N_{CN} shortly after it reached the peak value which can be attributed to significant variation of Aitken mode during the event. For example, the model failed to reproduce concentration trend of 10-60 nm particle at the early event stage (Fig. 10a-b). For the December 12 event, the model underpredicted a significantly lower peak concentration (about 4100 cm^{-3} lower) at about 12:00 pm than the measured one, due probably to presence of a significant amount of larger background particles (100-200 nm) which were not taken into account in the model (Fig. 9c and Fig. 10c). As a result, the N_{CCN} was underpredicted in two Heshan events (fig. 10a- b), owing to the fluctuation of background particle distribution and unexplained increase of concentration of particles at a size range of 10-60 nm at the beginning of event."

(1) In section 3.3, we mainly discussed the relationship between the dynamic processes and the N_{CCN}. We found that both the NPF characteristics and the properties of newly-formed particles could influence the N_{CCN}. We added several sentences to discuss the impact factors on the N_{CCN} in lines 501-514,

"To compare different impacts of the characteristics and properties of newly-formed particles, the N_{CCN} was simulated through varying parameters of different characteristics (case 1, 4 and 7) and properties (case 2, 3, 5, 6, 8 and 9). The input parameters for different cases are shown in Table S1. For case 2, 3, 5, 6, 8 and 9 scenarios, the surface tension or hygroscopicity was adjusted to match similar N_{CCN} values based on different NPF characteristics (case 1, 4 and 7, respectively). The results show that doubling GR produces the most significant impact on the N_{CCN}, and the surface tension (κ value) was adjusted to 0.030 N m^{-1}(1.2) to have the same impact (Fig. 8a). Obviously, a κ value of 1.2 for hygroscopicity is much higher than that of many inorganics, e.g., H_2SO_4 (κ=0.90, Topping et al., 2005) and NH_4NO_3 (0.58, Topping et al., 2005). Meanwhile, the surface tension was lower than the values (0.049-0.060) reported previously (Ovadnevaite et al., 2017; Engelhart et al., 2008; Cai et al., 2018). However, doubling GR value (16.0 nm h^{-1}) was reasonable and consistent with previous studies (Mönkkönen et al., 2005; Foucart et al., 2018; O'Dowd et al., 1999), suggesting significant contribution of GR to the growth. For doubling formation rate and halving PNSD, the modified surface tension and κ values were minor (Fig. 8b and c)."

Table S1. The input parameters for Case 1-9.

2 × and 0.5 × represent doubling and halving the parameters, respectively.

	GR	J	PNSD	$\sigma_{s/a}$ (N m^{-1})	κ
Case 1	2 ×	1 ×	1 ×	0.0728	Measured
Case 2	1 ×	1 ×	1 ×	0.030	Measured
Case 3	1 ×	1 ×	1 ×	0.0728	1.2
Case 4	1 ×	2 ×	1 ×	0.0728	Measured
Case 5	1 ×	1 ×	1 ×	0.065	Measured
Case 6	1 ×	1 ×	1 ×	0.0728	0.15
Case 7	1 ×	1 ×	0.5 ×	0.0728	Measured
Case 8	1 ×	1 ×	1 ×	0.067	Measured
Case 9	1 ×	1 ×	1 ×	0.0728	0.13

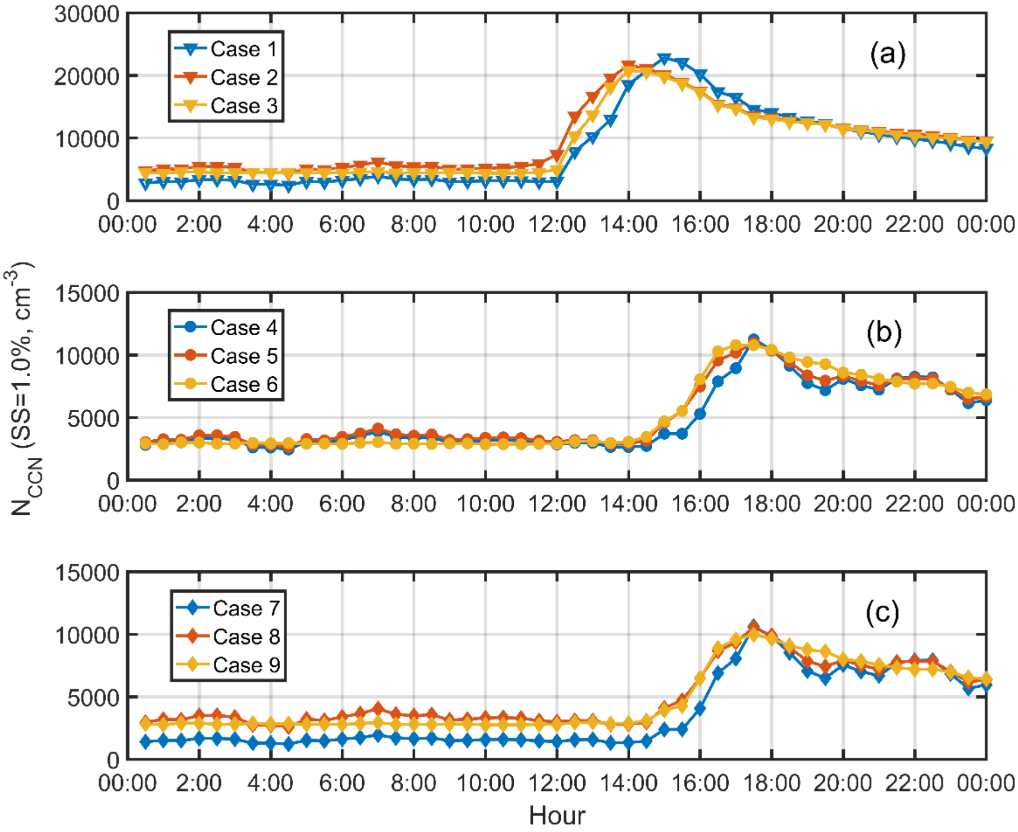

Figure. 8 The model N_{CCN} based on different characteristics (doubling growth rate and formation rate, and halving background particle distribution) and particle properties. Different colors and markers represent case 1-9, respectively.

”

5. With respect to surface tension, the authors are encouraged to review/include recent studies on the impact of morphology of organic/inorganic mixture on surface tension. As such, the discussion would be more in-depth and balanced.

Reply: We thank the reviewer for valuable suggestions. We have added discussion (also the references) on some recent studies to show the impact of liquid-liquid phase separation on surface tension and hygroscopicity in lines 352-362,

"This effect was closely related to the presence of liquid-liquid phase separation (LLPS) (Renbaum-Wolff et al., 2016), which was observed in organic-containing particles under high relative humidity. LLPS is mainly depended on the chemical composition of organics (e.g., functional groups and oxidation state) and inorganic-organic mixing ratio (Ruehl et al., 2016; Ma et al., 2021; Bertram et al., 2011). Once LLPS occurred, organic-rich phase on the droplet surface would reduce surface tension and further enhance water uptake (Rastak et al., 2017;Freedman, 2017). Surface tension is expected to increase with droplet growth, since the organic-rich phase becomes thinner and shifted to water-rich phase (Liu et al., 2018; Renbaum-Wolff et al., 2016; Ovadnevaite et al., 2017). Further laboratory and field studies are needed for better understanding the occurrence of LLPS in particles, its variation with different chemical composition, and its impact on the surface tension."

Minor comments:

1. A schematic diagram of the experimental setup is recommended.

Reply: We added a schematic diagram in section 2.1 and rephrased some sentences in lines 144-146: "Two aerosol sampling ports equipped respectively with a PM_{10} impactor and a $PM_{2.5}$ impactor were made of a 6 m long 3/8″ o.d. stainless-steel tube. The schematic diagram of the inlet system and instrument setup is shown in Fig. S1."

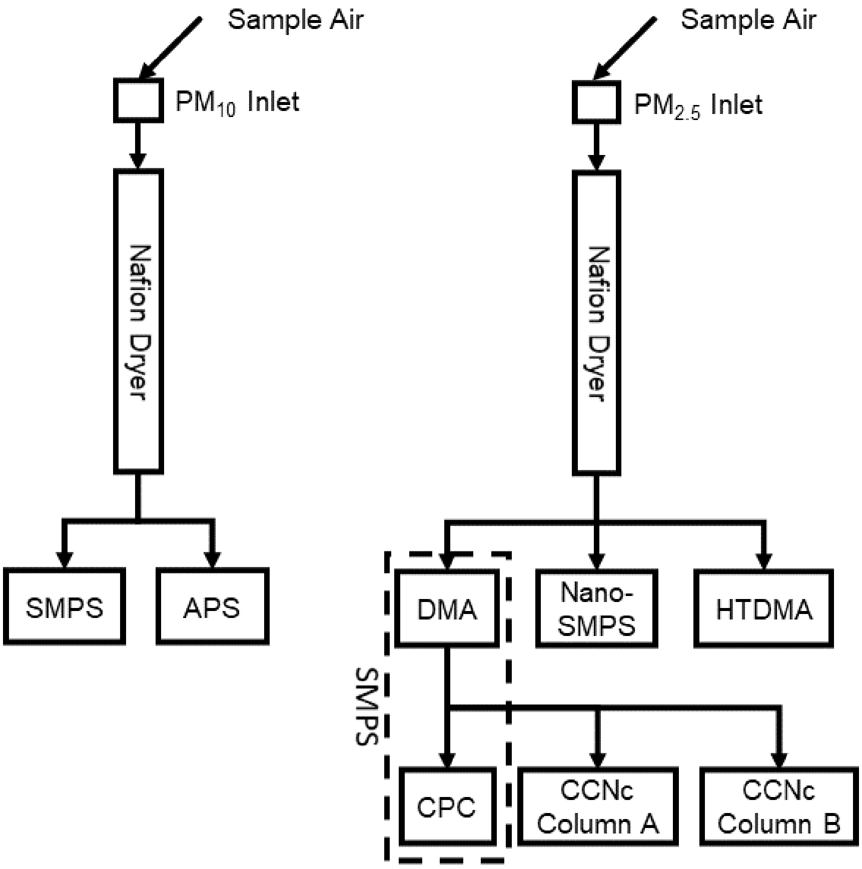

Figure S1. Schematic diagram of the experimental setup

2. The lowest measurable particle diameter in this study is 1 nm. Is there any reason not to use this for the estimation of formation and growth rates, instead of 3 nm (lines 227, 253, 265)?

Reply: The particle number size distribution (PNSD) data during the campaign was acquired by a commercial Nano-SMPS instrument. The instrument is controlled by Aerosol Instrument Manager (version 10, TSI Inc., USA) which does not provide accurate corrections for multiple charges and diffusion losses for particles smaller than 3 nm. While accurate inversion for particles smaller than 3 nm is still under development, we believe that it is adequate to use particles larger than 3 nm for modeling NPF in this study. Hence, we only used PNSD for particles larger than 3 nm to calculate formation and growth rates in this study. We added several sentences to clarify this issue in lines 159-163,
"The data inversion processes for the measured PNSD were done by Aerosol Instrument Manager (version 10, TSI Inc., USA). However, accurate inversion for particles smaller than 3 nm is currently still lacking due to large uncertainties from corrections for multiple charges and diffusion losses. Thus, we only discussed PNSD for particles larger than 3 nm in this study."

3. Line 415 and other instances, the "fail" is misspelled as "fell."
   Reply: Typos have been corrected in lines 436, 443 and 446.

Reference:

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