

**Anonymous Referee #1:** This manuscript investigates CCN production associated with atmospheric new particle formation using a few observed cases at a mountain-top measurement station. The scientific approach appears to be robust, and the paper has some novel features, such as introduction of the "time window" concept. Interpretation of the results requires, however, considerable revisions here and there. The language of the paper requires also improvements. My detailed comments in this regard are given below.

[Response] The authors sincerely thank the reviewer for the positive and constructive assessment, as well as for the thoughtful and thorough review. We appreciate the reviewer's recognition of the study's robust approach and novel features. In direct response to the general suggestion regarding language, the English throughout the entire manuscript has been thoroughly polished by a native-speaking expert to enhance clarity and readability. A detailed, point-by-point response to each specific comment follows below (in blue color), along with a description of the corresponding revisions made to the manuscript text (in red color). We hope that the revised manuscript, together with our clarifications, fully addresses all the concerns raised.

### Scientific issues

**Lines 29-32:** in Abstract: While changes in CCN concentrations influence many cloud properties via changes in cloud droplet concentrations, the ability of a cloud to form is not really dependent on CCN concentrations (unless CCN are missing altogether, which is extremely rarely the case). I recommend mentioning changes in cloud properties in this context, not talking about cloud formation.

[Response] We thank the reviewer for the suggestion, and the mentioned sentence has been revised as:

“Furthermore, the duration of NPF-to-CCN conversion was quantified using a ‘Time Window ( $\tau$ )’, revealing that polluted conditions accelerated the conversion by 17.0% ( $\tau = 16.4$  h vs. 19.8 h). Nitrate played an important role in maintaining a rapid particle growth rate, thereby shortening  $\tau$  and enhancing CCN production from NPF—a process that can ultimately influence cloud microphysical properties by increasing the potential cloud droplet number concentration.”

**Lines 218-219:** Talking about hygroscopic growth in this context is misleading. Being a CCN at a given supersaturation (SS) is an aerosol property, dictated by aerosol size and chemical composition. Becoming a CCN means increasing either particle “dry” size (by condensation growth) or its hygroscopicity to a sufficient degree. This should not be mixed with particle hygroscopic growth which happens when CCN respond to ambient RH or are activating into cloud droplets.

[Response] We thank the reviewer for pointing out this critical clarification, and the mentioned sentence has been revised as:

“This approach more clearly describes the dynamic process in which newly formed particles

grow via condensation (increasing dry size and/or altering chemical composition) to the critical size and hygroscopicity required to act as CCN at defined supersaturation, and thus extends current methodologies by offering a more process-explicit framework to evaluate how precursor conditions and chemical pathways modulate the climatic impact of NPF.”

**Lines 311-313:** The logic of this statement does not work. The growth rate of a newly formed particle is determined solely by the condensation flux of low-volatile (and partly semi-volatile) compounds into it, being influenced mainly by the gas-phase concentration of such compounds. To a first approximation, the hygroscopicity of the condensing compounds do not matter in this context, as growth rates are usually determined for “dry” particles or for particles at low RH. It is true that the hygroscopicity of the condensing material influences the size of a growing particles at elevated RH, so that particles having more hygroscopic material have a higher “wet” size, which enhances the condensation flux and thereby growth rate of particles to some extent. But this a secondary effect compared with gas-phase concentrations.

[Response] We thank the reviewer for pointing out this important issue. We now clarify it as follow:

“Our findings indicate that in anthropogenically influenced mountain regions, nitrate—primarily as ammonium nitrate ( $\text{NH}_4\text{NO}_3$ )—can serve as a competitive source of low-volatility condensable vapor, partially substituting for organics in driving the mass growth of new particles. This occurs under conditions of elevated  $\text{NO}_2$  and  $\text{NH}_3$ , where efficient photochemical production and gas-to-particle partitioning of  $\text{NH}_4\text{NO}_3$  are favored. While the strong hygroscopicity of nitrate plays a secondary role by increasing the particle’s wet size (and thus potentially enhancing condensation efficiency under high relative humidity), its primary contribution to growth is through direct vapor condensation.”

**Lines 326-327:** Besides these theoretical calculations and laboratory experiments, there is also evidence from field measurements on the involvement of ammonia/amines. I recommend citing also field evidence in this context.

[Response] We thank the reviewer for the suggestion. The related references are cited here.

“Previous field and chamber studies also proposed that the gaseous species such as ammonia (Kulmala et al., 2013; Kürten et al., 2019) and amines (Metzger et al., 2010; Yao et al., 2018) also promote the nucleation.”

**Line 359:** Was there some specific reason for why only a single SS (0.2%) was used in the calculations. Having two or even more values of SS in consideration may have given additional information on how newly formed particle reach CCN sizes. This is especially so when noting that with typical particle growth rates, it takes quite a while until newly formed particle grow into sizes relevant for CCN at 0.2% (at higher values of SS, which are certainly possible, newly formed particles become CCN much quicker).

[Response] We thank the reviewer for this insightful and constructive suggestion regarding

the consideration of multiple supersaturation (SS) levels. We agree with the reviewer that analyzing CCN activation at different SS values provides valuable information on the timescale and efficiency with which newly formed particles grow into CCN. Our initial focus on a single SS (0.2%) was primarily for clarity in presenting the core comparison between event types and for consistency with a referenced regional SS measurement (Gong et al., 2023). However, we fully agree that a multi-SS analysis significantly enriches the discussion. We now present the critical activation diameter ( $D_a$ ) and the resultant CCN number concentration ( $N_{CCN}$ ) for two representative SS values: 0.2% (representing a common in-cloud condition) and 0.4% (representing a higher, yet plausible, supersaturation). This dual-SS approach allows us to discuss how the required growth size and the subsequent CCN production potential vary with SS. The analysis confirms that at higher SS (0.4%), particles activate at a smaller  $D_a$ , leading to a shorter theoretical growth timescale ( $\tau$ ) and a higher instantaneous  $N_{CCN}$  from the growing nucleation mode. In addition, we have revised this part in section 3.4 accordingly.

“To quantify the CCN production from NPF events, the  $N_{CCN}$  was calculated. Since supersaturation (SS) cannot be measured directly at the site, we employed a sensitivity approach using two representative SS values. These values were selected based on prior aircraft measurements in the regional background atmosphere, which reported a range of 0.1-0.5% (Gong et al., 2023). We performed calculations for SS=0.2% and SS=0.4%, encompassing a common in-cloud condition and a higher activation threshold. For each SS, the critical activation diameter ( $D_a$ ) was derived using  $\kappa$ -Köhler theory, with the hygroscopicity parameter ( $\kappa$ ) estimated from the measured particle chemical composition (Bougiatioti et al., 2011), adjusting for local altitude. The calculated  $N_{CCN}$  for both SS levels was then compared with observed NCCN to evaluate the parameterization's performance and to analyze the SS-dependence of CCN production efficiency.”

“The critical diameter for CCN activation ( $D_a$ ) exhibited a strong dependence on supersaturation, as theoretically expected. For the studied NPF events,  $D_a$  at SS=0.4% was substantially lower than at SS=0.2%. Under the lower SS condition (0.2%),  $D_a$  varied from 111 to 129 nm, with a higher average in polluted (NPF-P) events (126 nm) compared to clean (NPF-C) events (120 nm). This difference correlated with a lower average hygroscopicity parameter ( $\kappa$ ) for NPF-P events (0.18) than for NPF-C events (0.21), originating from a higher organic mass fraction (76.6% vs. 65.4%). At the higher SS of 0.4%, the average  $D_a$  decreased to approximately 80 nm (NPF-P) and 76 nm (NPF-C), yet the inverse relationship between  $D_a$  and  $\kappa$  persisted.

The dependence of  $D_a$  on supersaturation has significant implications for NPF-driven CCN production. At a higher SS of 0.4%, the substantially reduced critical diameter shortens the required growth trajectory and timescale, allowing particles to become CCN-active more

rapidly in environments with elevated supersaturation. Consequently, the net CCN enhancement ( $EF_{CCN}$ ) during NPF was systematically greater at  $SS=0.4\%$  than at  $0.2\%$ . Notably, while pollution-enhanced CCN production was evident at both SS levels, the relative enhancement of NPF-P over NPF-C events was more pronounced at the lower SS ( $0.2\%$ ). This indicates that the chemically processed, faster-growing particles in polluted air masses are particularly effective at overcoming the greater activation barrier (larger  $D_a$ ) at low SS.”

**Lines 362-364:** The logic of this statement does not work either. It is true that for 2 particles of similar size, the one with a higher organic fraction requires a higher SS to activate into a cloud droplet (as organics tend to have much lower hygroscopicity than the main inorganic compounds). However, a higher organic fraction is not expected to suppress condensation of other material from the gas phase (not even water unless these organics reduce e.g. the accommodation coefficients of condensing water). Please modify.

[Response] We thank the reviewer for pointing this out. We now modify it in the revised version according to your suggestion.

“This difference correlated with a lower average hygroscopicity parameter ( $\kappa$ ) for NPF-P events ( $0.18$ ) than for NPF-C events ( $0.21$ ), originating from a higher organic mass fraction ( $77\%$  vs.  $65\%$ ).”

**Lines 390-393:** It is unclear to me how the degree of oxidation of organics can be estimated from VRF in mixtures of inorganic and organic compounds?

[Response] We thank the reviewer for pointing this out. We now provided more discussion in the revised version to clarify this issue.

“The VFR in the  $14\text{-}80\text{ nm}$  size range was  $10\text{-}20\%$  (Fig. 4d), significantly higher than values reported for polluted urban Beijing ( $\sim 5\%$ ; Wu et al., 2017). Because heating to  $300\text{ }^{\circ}\text{C}$  effectively removes volatile inorganic salts and semi-volatile organic compounds, a higher VFR primarily reflects a greater abundance of low-volatility organic compounds (LVOCs). At our background site, where local combustion influence is minimal, this points to a more aged, oxidized organic aerosol component (Ehn et al., 2014; Jimenez et al., 2009), consistent with the observed lower  $\kappa$  and higher  $D_a$ .”

**Lines 505-507:** There is something strange in this sentence. I am able to understand what is meant to be said.

[Response] We thank the reviewer for pointing this out. We now modify it in the revised version.

“Collectively, these results suggest that cross-regional pollutant transport enriches precursor concentrations, elevates the atmospheric oxidation capacity, and thereby enhances both the magnitude and advances the timing of CCN production at the boundary layer top. Crucially, while previous studies have indicated that intense local pollution can suppress CCN formation from NPF, our findings demonstrate that in oxidizing, transport-influenced environments such

as the one studied here, aged pollution plumes can instead amplify CCN yields. Accurately representing these oxidation-driven growth pathways in atmospheric models is therefore essential for constraining aerosol-cloud-climate feedbacks in rapidly developing regions.”

### Technical issues

1. The paper has several typos, especially in section 3, which should be corrected in the revised version. Just a few examples of these: (line 231: event is, line 247: particles, line 248: the in-cloud, line 282: which is close to, line 296: GR values are higher by, lines 402-403: non-volatile)

[Response] We sincerely thank the reviewer for this meticulous and helpful feedback. We have carefully addressed all the specific typographical errors and similar issues identified in Section 3 and throughout the manuscript. The detailed corrections are as follows:

Line 231: Corrected "event are" with "event is"

Line 247: Corrected "newly formed particle" with "newly formed particles".

Line 248: The previous sentence has now been revised.

Line 282: The sentence has been revised to: “Our values are close to those reported for other Chinese high-altitude background sites like Mount Tai ( $J_3 = 1-2 \text{ cm}^{-3} \text{ s}^{-1}$ ; Shen et al., 2019) ...”

Line 296: The sentence has been revised to: “Compared to typical values reported for a remote boreal forest site (Hyytiälä, Finland:  $J_3 = 0.4 \text{ cm}^{-3} \text{ s}^{-1}$ , GR = 2.3 nm h<sup>-1</sup>; Kerminen et al., 2018), the formation and growth rates observed at our site are higher by 275% and 126%, respectively.”

Lines 402-403: Corrected "no-volatile" with "non-volatile".

We have conducted a thorough proofreading of the entire manuscript to eliminate any remaining typographical or grammatical errors.

2. The percentages given in the paper appear overly accurate (one digit, e.g. lines 289, 292 and 294, but also elsewhere). Maybe 1% accuracy would be more relevant here.

[Response] We thank the reviewer for this insightful comment regarding the appropriate level of precision for reporting percentages. In response, we have systematically revised all quantitative results throughout the manuscript, including those on lines 289, 292, and 294, by rounding to the nearest whole number (e.g., from 23.2% to 23%, from 39.2% to 39%, from 23.6% to 24%).

3. A similar accuracy issue concerns critical diameters in lines 360-362 (4 digits too many).

[Response] We thank the reviewer's suggestion. The critical diameters are now reported

as integers, with all extraneous decimal places removed.

4. All the figures should be understandable based on available figure legends and figure captions, and the figures should be consistent with what is said in the text. This is not the case for many of the figures:

*[Response]* We sincerely thank the reviewer for highlighting this critical issue regarding the clarity and consistency of our figures. We have carefully reviewed each figure in the manuscript and implemented the comprehensive revisions to fully address your concern. First, we have thoroughly revised all figure captions to provide a complete, self-contained description of the data shown, including the specific conditions, parameters, and conclusions that can be drawn directly from the figure. All abbreviations and technical terms used in the figures are now clearly defined. Second, we have performed a line-by-line cross-check between the manuscript text and each figure. All descriptions, interpretations, and numerical references to the figures have been verified and, where necessary, corrected to ensure they accurately reflect the visual data presented.

5. In Figure 1, it is not mentioned which one (a or b) corresponds to clean or polluted conditions (I suppose a is clean and b is polluted based on the distributions).

*[Response]* We thank the reviewer for this precise and helpful feedback regarding Figure 1. We have revised the figure caption accordingly to explicitly state the conditions represented in each panel. The revised caption now reads:

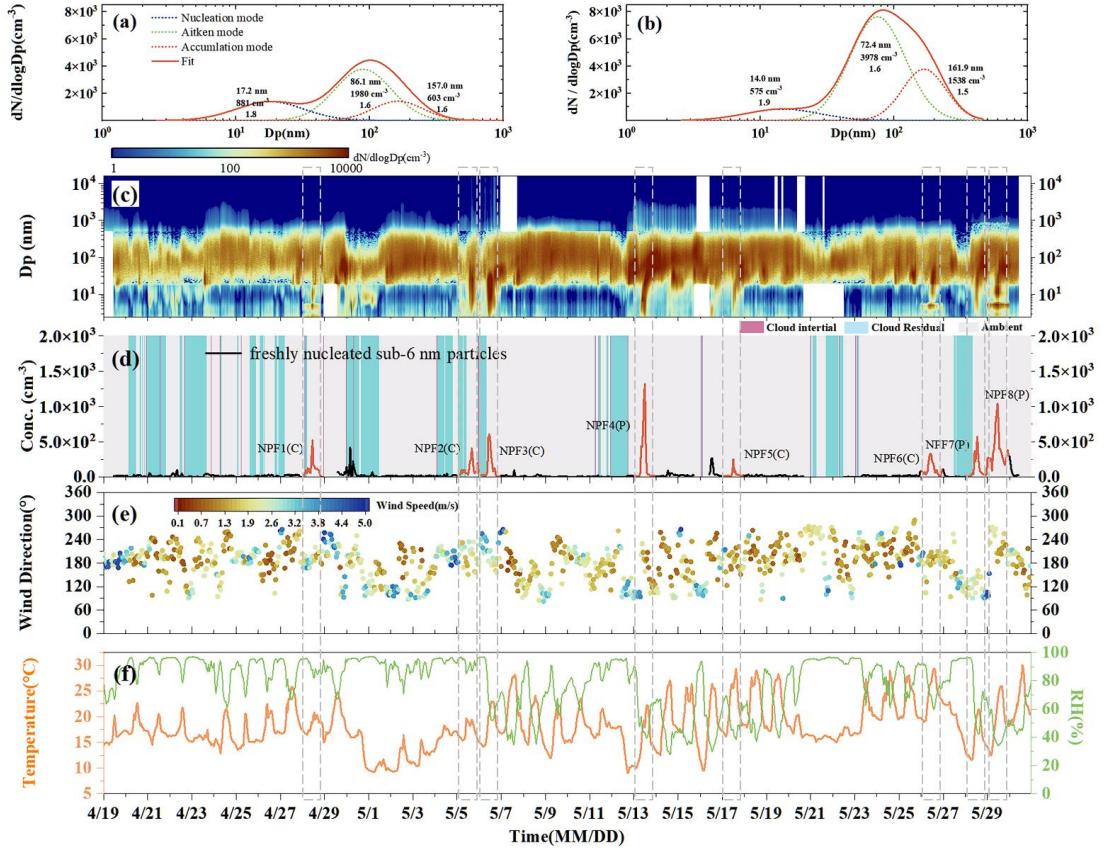


Figure 1. Overview of atmospheric conditions and new particle formation (NPF) events at the mountain-top station. The dashed-line frame represents the NPF event days. (a-b) Lognormal-fitted particle number size distributions for representative (a) clean (NPF-C) and (b) polluted (NPF-P) NPF events. Fitted modes are color-coded: nucleation (<20nm, blue), Aitken (20-100nm, green), and accumulation (100-1000nm, orange). (c) Time series of observed particle number size distributions ( $dN/d\log D_p$ ) during the entire campaign. (d) Temporal evolution of particle types: cloud interstitial (dark red), cloud residual (light blue), and non-cloud periods (Ambient, light gray). The occurrence of sub-6nm particles (fresh nucleation) is overlaid as red lines, highlighting identified NPF event days. (e) Wind direction time series, where color intensity represents wind speed magnitude. (f) Time series of temperature and relative humidity.

6. In Figure 2g, it is not explained which bars represent J and which ones GR (this info can now only be gotten by reading the main text).

[Response] We thank the reviewer for this precise and helpful feedback regarding the clarity of Figure 2g. We have revised the figure legend and the corresponding figure element to explicitly differentiate the data series. The updated figure now clearly labels the bars representing the nucleation rate (J) and those representing the growth rate (GR), ensuring the information is directly accessible from the figure itself. In addition, we also revised the figure caption accordingly to explicitly state the conditions represented in each panel. The revised caption now reads:

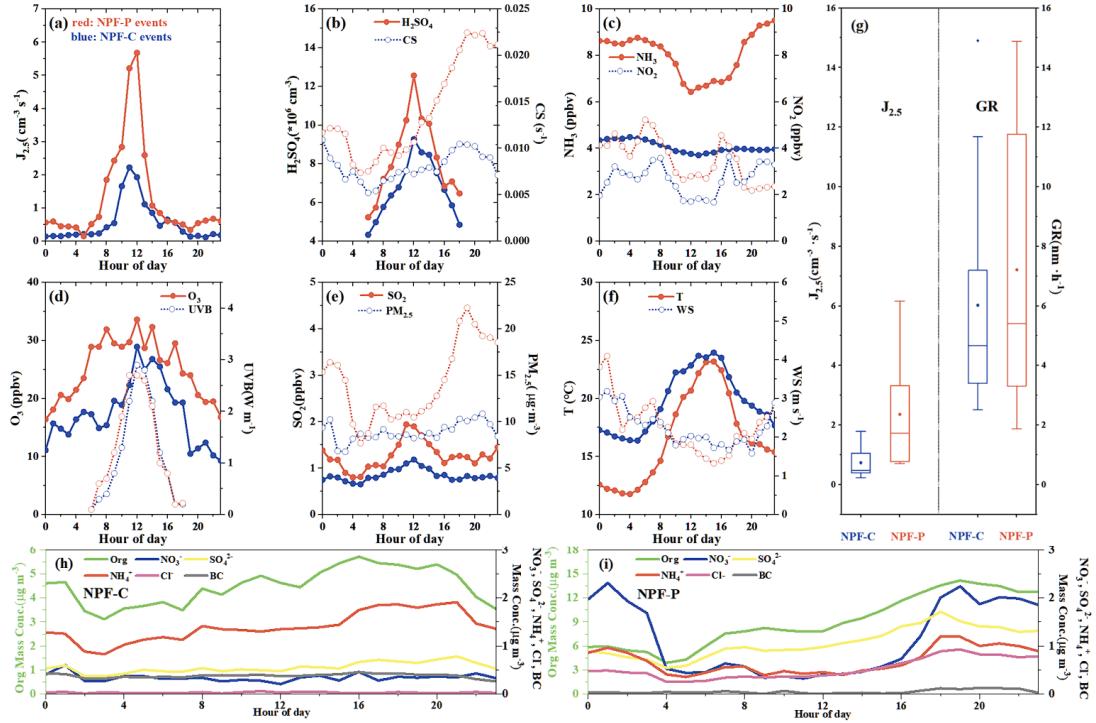


Figure 2: Diurnal comparison of key parameters and new particle formation (NPF) metrics between clean (NPF-C) and polluted (NPF-P) event days. (a) formation rate ( $J_{2.5}$ ); (b)  $\text{H}_2\text{SO}_4$  concentration and condensation sink (CS); (c)  $\text{NH}_3$  and  $\text{NO}_2$  concentration (d)  $\text{O}_3$  concentrations and UV-B radiation intensity; (e)  $\text{SO}_2$  concentration and  $\text{PM}_{2.5}$  mass concentration; (f) Temperature (T) and wind speed (WS); (g) Box plots of formation rate ( $J_{2.5}$ ) and growth rate (GR), where boxes show the interquartile range (25th-75th percentile), internal lines denote the median, dots represent the arithmetic mean, and whiskers extend to the 10th and 90th percentiles. (h-i) Mean diurnal profiles of non-refractory  $\text{PM}_{2.5}$  chemical composition (organics, sulfate, nitrate, ammonium, chloride) and black carbon (BC) mass concentration for (h) NPF-C and (i) NPF-P events.

7. The text says (lines 345-346) that Figs. 3b and 3c give  $J$  as a function of SA and ammonia product, but this is not true for Fig. 3b.

[Response] We thank the reviewer for the careful correction. We have revised the relevant text in lines 345-346 to accurately reflect the content of the figures:

“A pronounced linear relationship exists between  $J_{2.5}$  and the product of  $\text{H}_2\text{SO}_4$  and  $\text{NH}_3$  concentrations (Fig. 3c). The Pearson correlation coefficient ( $R$ ) for  $J_{2.5}$  versus  $[\text{H}_2\text{SO}_4] \times [\text{NH}_3]$  ranges from 0.79 to 0.92, notably higher than the correlation of  $J_{2.5}$  with  $[\text{H}_2\text{SO}_4]$  alone ( $R = 0.77\text{-}0.87$ , Fig. 3b). ”

8. The text says (lines 373-374) that Figs. 4b reveals something about the coupling between critical diameter and temporal particle size evolution. This is practically impossible to see from Figure 4b alone, but it requires additional information from other figures and text.

[Response] We sincerely thank the reviewer for this important observation. We admit that the coupling between critical diameter ( $D_a$ ) and temporal particle size evolution cannot be directly discerned from Figure 4b alone, as this panel primarily presents the temporal evolution of  $N_{CCN}$  and its activation ratio (AR). The coupling analysis is in fact based on the combined interpretation of Figure 4a (which shows the time-series of  $D_a$ ) together with the particle size distribution data presented in Figure 1c and the growth rate information in Figure 2. We have therefore revised the text and added Figure S4 in Supplementary Information to clarify this issue.

“The diurnal evolution of the particle population further elucidates the transition from nucleation to CCN production. Total particle number concentration ( $N_{CN}$ ) begins a rapid increase after ~07:00 LT, driven by the nucleation burst (Fig. 4b). Although CCN concentration ( $N_{CCN}$ ) starts to rise concurrently, the explosive production of small nucleation mode particles initially causes the activation ratio ( $AR = N_{CCN}/N_{CN}$ ) to decline, reflecting the time required for growth to CCN active sizes.  $N_{CCN}$  subsequently peaks around 09:00–10:00 LT, approximately 2–3 hours after the  $N_{CN}$  surge, marking the period when a substantial fraction of newly formed particles has grown sufficiently. After ~14:00 LT, as growth processes intensify (indicated by high GR), an increasing number of particles reach  $D_a$ , and the AR begins a gradual recovery (Figs. S4b-c).”

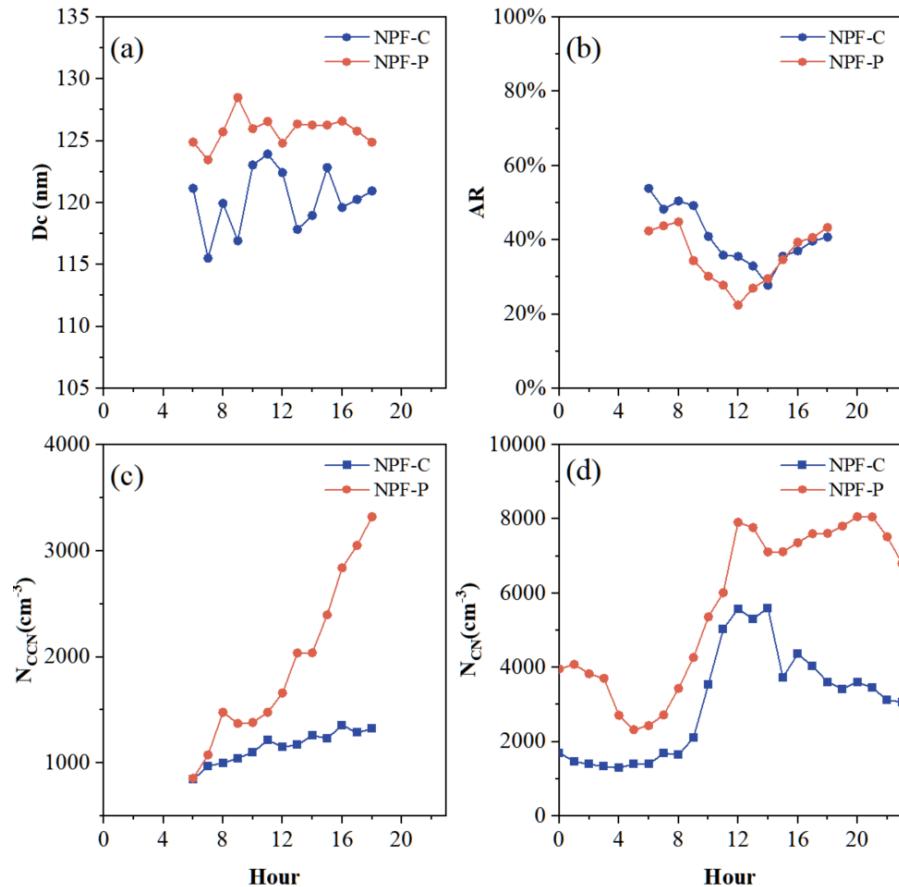


Figure S4: The diurnal variation of critical activation diameter ( $D_a$ ), activation ratio (AR),

the number of cloud condensation nuclei ( $N_{CCN}$ ) and total particle number concentration ( $N_{CN}$ ) in NPF-C and NPF-P events. The blue line denotes to NPF-C events and red line denotes to NPF-P events.

9. In Figure 5b, the 3 red cases seem to refer to polluted ones, while the blue cases correspond to clean ones. This is not explained in figure caption, which is confusing especially as red and blue mean totally different things in Figure 5a.

[Response] We sincerely thank the reviewer for this careful and constructive comment. We have now thoroughly revised the caption for Figure 5 to provide a complete and unambiguous description for each panel.

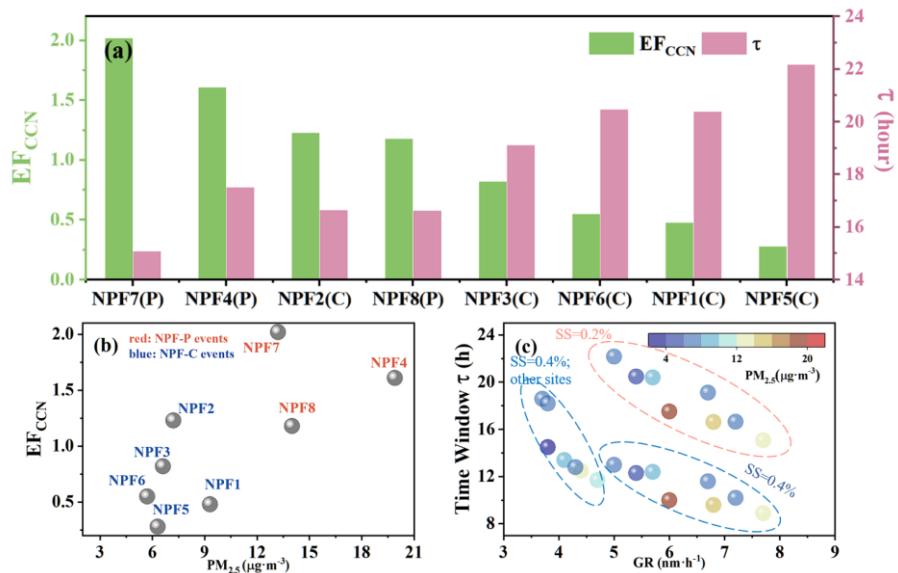


Figure 5: Relationships between CCN enhancement factors, Time Window ( $\tau$ ), and environmental parameters. (a) Scatter plot of the CCN enhancement factor ( $EF_{CCN}$ ) versus the Time Window ( $\tau$ ) for particle growth to CCN size across all eight NPF events. (b) Correlation between the  $PM_{2.5}$  mass concentration and  $EF_{CCN}$ , where individual data points are color-coded to distinguish between NPF events occurring under polluted (red font) and clean (blue font) conditions. (c) Relationship between the particle growth rate (GR) and  $\tau$ . The color gradient represents the concurrent  $PM_{2.5}$  mass concentration at the Shanghuang station for data at supersaturations of 0.2% and 0.4%. Data from other sites (shown for SS=0.4%) are included for comparison.

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