

# Reply to comments

Journal: Atmospheric Chemistry and Physics

Manuscript Number: acp-2022-440

Title: "High frequency of new particle formation events driven by summer monsoon in the central Tibetan Plateau, China"

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## I. Reply to Reviewer 1

### Reply to Reviewer 1's overall comments:

*This manuscript investigates atmospheric new particle formation (NPF) taking place in the central Tibetan Plateau. To my knowledge, there are no prior publications of NPF in this location, so the obtained results can be considered worth publishing. The paper itself is well organized and the conducted analysis appears to be scientifically sound. There are, however, two major issues that require further consideration.*

We appreciate the comments from the reviewer on this manuscript. We have answered them point to point in the following paragraphs (the texts italicized are the comments, the texts indented are the responses, and the texts in blue are revised parts in new manuscript). In addition, all changes made are marked in the revised manuscript. Thanks for the reviewer's affirmation on our work.

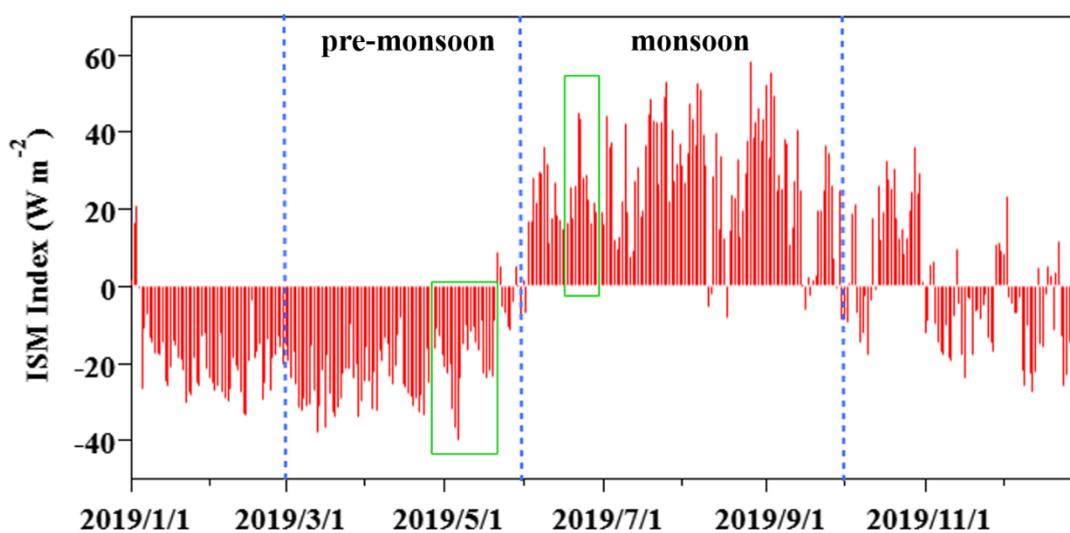
### Reply to Reviewer 1's major comments (2):

*1. First, the measurements periods are rather short, about 4 weeks for the pre-monsoon season and less than 2 weeks for the monsoon season. As a result, it remains unclear how representative the obtained results are for this location during these two seasons.*

Thanks for the comment. The measurements periods were a little limited as the reviewer described. But our measurements periods can be representative for this location during pre-monsoon season and monsoon season as follows:

1) The intensity of Indian Summer Monsoon during the two measurements periods can represent that in the whole pre-monsoon and monsoon seasons, respectively. The intensity of Indian Summer Monsoon is an important indicator to distinguish the monsoon season. Here the intensity of Indian Summer Monsoon

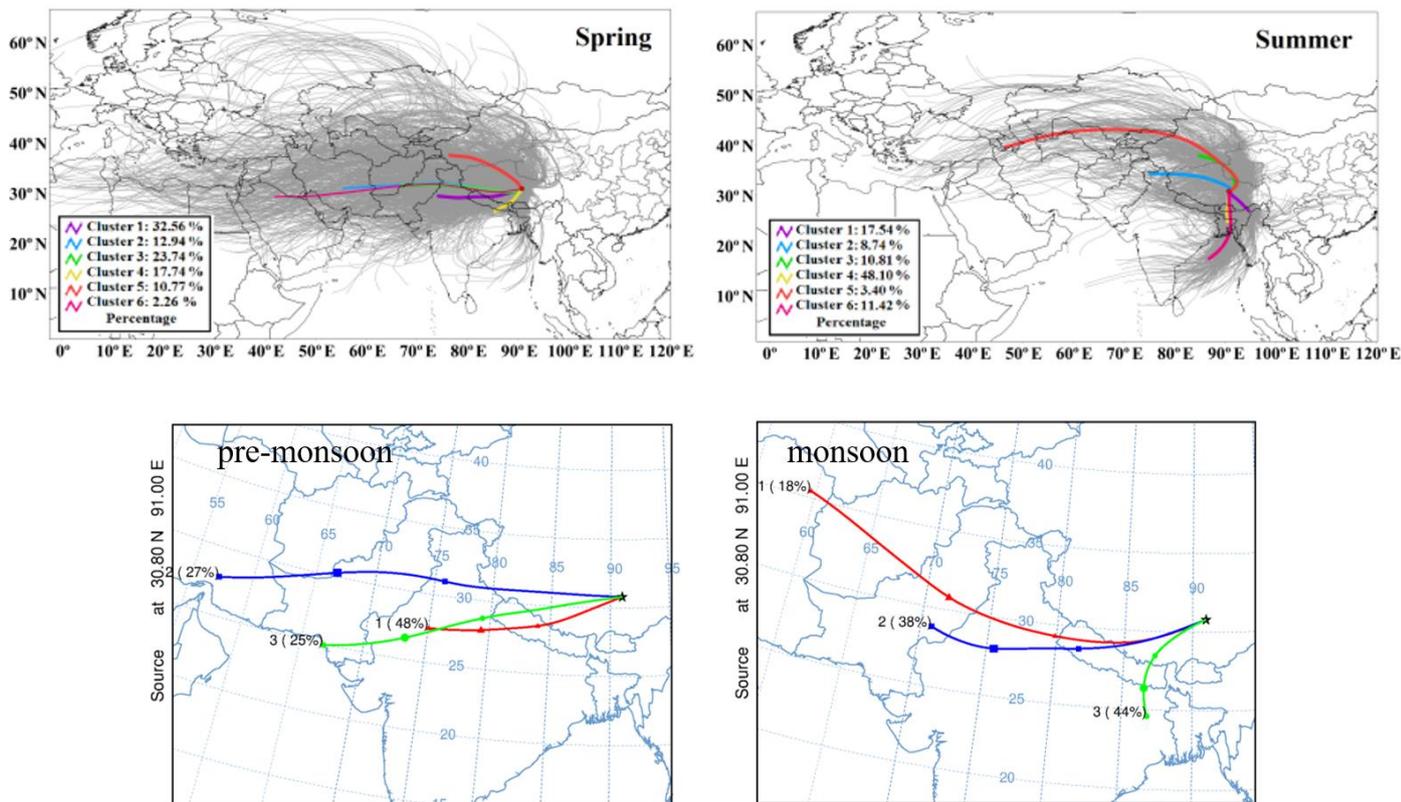
(ISM) was indicated by the ISM Index, which are defined by the negative outgoing longwave radiation anomalies (with respect to the climatological annual cycle) averaged over the Bay of Bengal–India region (10°–25°N, 70°–100°E) (Wang and Fan, 1999). As shown in Fig. R1, the measurement periods (green boxes) were in the pre-monsoon season (March-May) and monsoon season (June-September), respectively. And the IMS index during the two measurements periods were equivalent to those of the whole pre-monsoon season (average: -19.5 vs -20.7 W m<sup>-2</sup>) and monsoon season (average: 27.0 vs 26.3 W m<sup>-2</sup>), respectively. Therefore, we considered that these two observation periods are representative in the seasonal characteristics in pre-monsoon season and monsoon season, respectively.



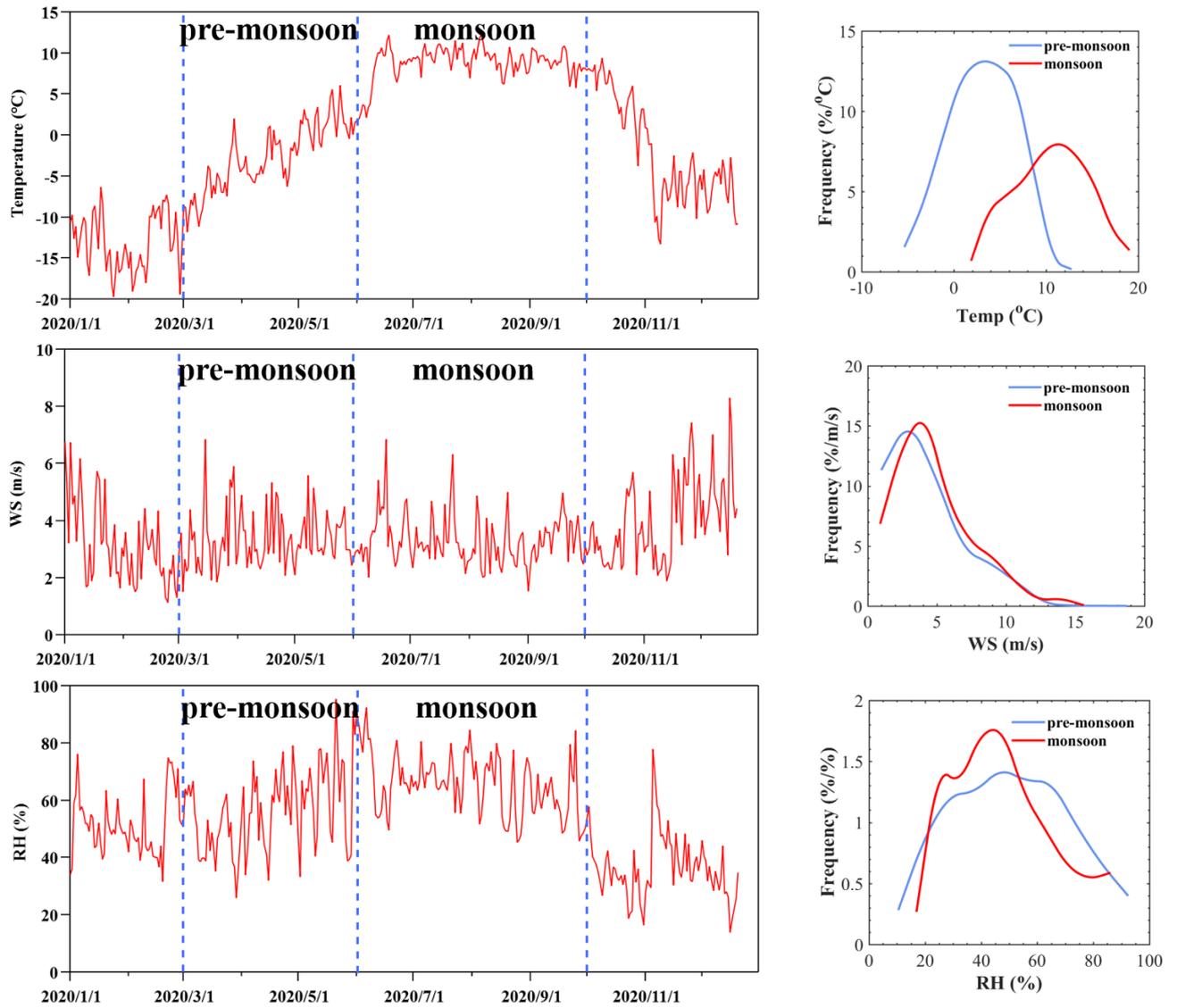
**Figure R1.** The Indian Summer Monsoon (ISM) Index in 2019. The measurements periods are marked by the green boxes.

2) The characteristics of meteorology and atmospheric pollutants in the two measurements periods was generally in agreement with the previous long-term studies at Nam Co station and other sites in the Tibetan Plateau (TP) (Yin et al., 2017; Cong et al., 2015; Bonasoni et al., 2010; Xu et al., 2018). Both previous research and this study showed that, strong westerlies pass through western Nepal, northwest India and Pakistan in pre-monsoon season, while air masses were mainly derived from Bangladesh and northeast India and brought moisture that originated in the Bay of Bengal in monsoon season (Fig. R2) (Yin et al., 2017). The temperature, WS and RH in the two measurements periods were matched with those in the whole pre-monsoon and monsoon season of 2020 at Nam Co station (Fig. R3) (National Tibetan Plateau Data Center). The average temperature in pre-monsoon and monsoon seasons were around 3 and 10 °C, respectively. WS showed no difference between pre-monsoon and monsoon seasons with the average value of 4 m/s. The average level of RH was similar between the two seasons, but the variation range of RH was larger in pre-monsoon season. In addition, the level of PM, BC and ozone in the two

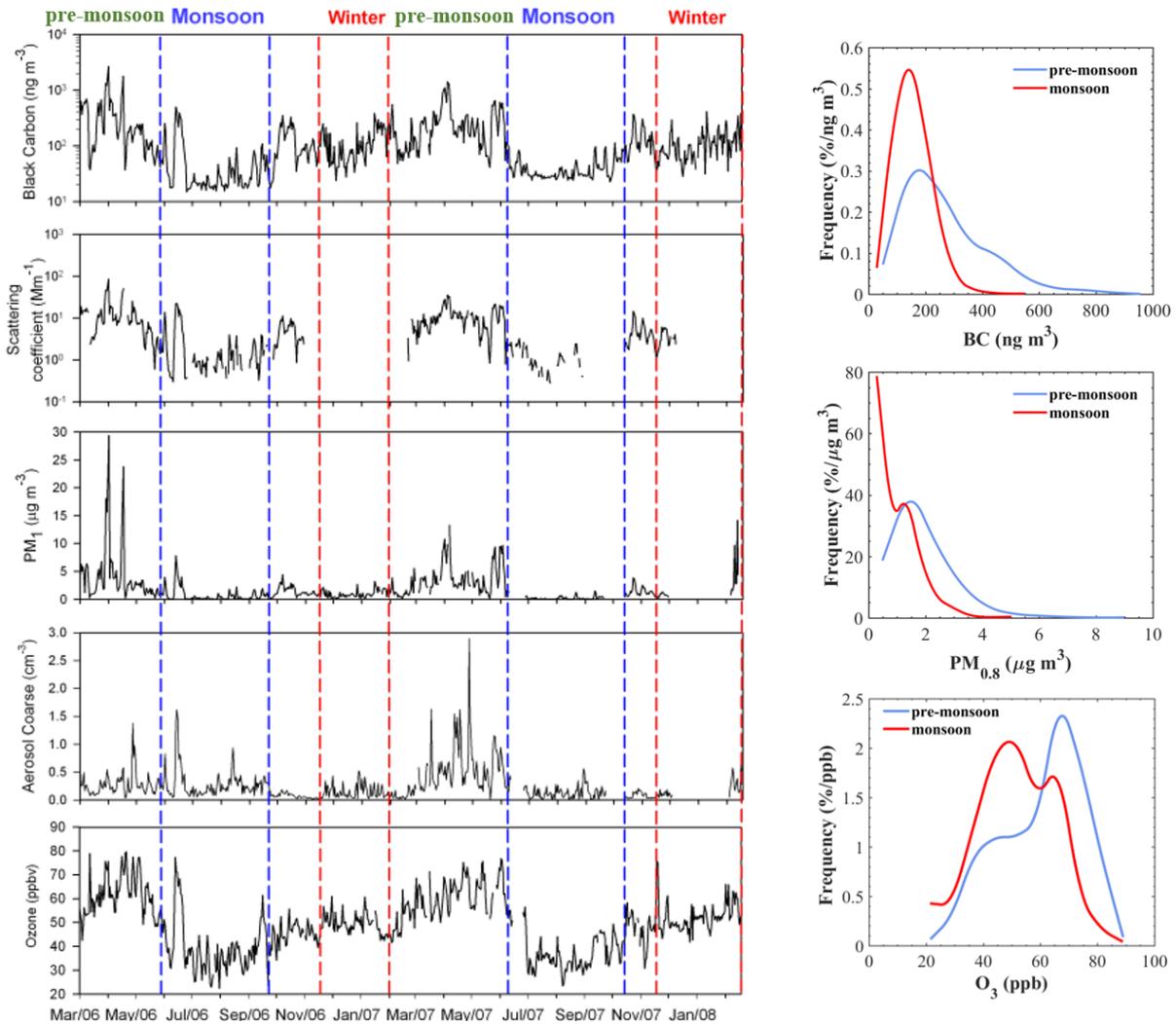
54 measurements periods were matched with those in the whole pre-monsoon and monsoon season at the  
 55 other site of the TP (Fig. R4) (Bonasoni et al., 2010; Xu et al., 2018). The average concentrations of PM<sub>1</sub>  
 56 (PM<sub>0.8</sub>) in pre-monsoon and monsoon seasons were around 1 and 2 μg/m<sup>3</sup>, respectively. The concentration  
 57 of BC was at a level of hundreds nanogram per cubic meter in pre-monsoon season, while at a lower level  
 58 in monsoon season. Ozone showed the similar pattern with BC. It should be noted that there will be some  
 59 differences between this study and other results due to the differences in time resolution. In a word, the  
 60 characteristics of meteorology and atmospheric pollutants in the two measurements periods in this study  
 61 can well reflect those in pre-monsoon and monsoon seasons.



62 **Figure R2.** Comparison of trajectories between this study and previous study at Nam Co station.  
 63 Backward HYSPLIT trajectories for each measurement day (black lines in the maps), and mean back  
 64 trajectory for six HYSPLIT clusters (colored lines in the maps) arriving at Nam Co Station in spring  
 65 (MAM) and summer (JJA) (Yin et al., 2017) (top). The frequencies of the 48 h back trajectories of air  
 66 masses arriving at Nam Co station from different directions during pre-monsoon and monsoon seasons in  
 67 this study (bottom).  
 68



69  
70 **Figure R3.** Comparison of meteorology between this study and the whole seasons in 2020. Time series  
71 of ambient temperature, wind speed and relative humidity at Nam Co station from January 2020 to  
72 December 2020 (National Tibetan Plateau Data Center) (left). Comparison in frequency distributions of  
73 temperature, WS and RH at Nam Co station in pre-monsoon and monsoon seasons in this study (right).



**Figure R4.** Comparison of atmospheric pollutants between this study and previous TP studies. Time series of f BC, aerosol scattering coefficient, PM<sub>1</sub>, coarse particle number and surface ozone at the Nepal Climate Observatory-Pyramid (NCO-P) station from 1 March 2006 to 28 February 2008 (Bonasoni et al., 2010) (left). Comparison in frequency distributions of BC, PM<sub>0.8</sub> and O<sub>3</sub> at Nam Co station in pre-monsoon and monsoon seasons in this study (right).

3) Based on the above discussion, the two measurements periods in this study can respectively represent the pre-monsoon and monsoon seasons at Nam Co station, so the NPF characteristics of the two observation periods can also be considered as the NPF characteristics in pre-monsoon and monsoon seasons. In addition, the phenomenon of higher NPF frequency in monsoon season than pre-monsoon season was also found in the other sites in the Tibetan Plateau (TP). A 16-month measurements from 2006 to 2007 at Himalayan Nepal Climate Observatory at Pyramid (NCO-P) site on the southern TP showed NPF frequency of 38% in pre-monsoon season and 57% in monsoon season (Venzac et al., 2008). At Mt. Yulong on the southeastern TP, the NPF frequency was only 14% during pre-monsoon season (Shang et al., 2018). The NPF frequency of 15% in pre-monsoon season and 80% in monsoon season at Nam Co

station was consistent with these studies, with more significant seasonal differences. The significant seasonal differences may be due to the fact that the occurrence of NPF is more sensitive to the monsoon in extremely clean background areas (such as Nam Co station and Mt. Yulong). In summary, our study emphasized the seasonal differences in NPF frequencies at Nam Co station, and the results was reliable.

Wang, B. and Fan, Z.: Choice of South Asian Summer Monsoon Indices, *Bulletin of the American Meteorological Society*, 80, 629-638, 10.1175/1520-0477(1999)080<0629:COASASM>2.0.CO;2, 1999.

Yin, X., Kang, S., de Foy, B., Cong, Z., Luo, J., Zhang, L., Ma, Y., Zhang, G., Rupakheti, D., and Zhang, Q.: Surface ozone at Nam Co in the inland Tibetan Plateau: variation, synthesis comparison and regional representativeness, *Atmos. Chem. Phys.*, 17, 11293-11311, 10.5194/acp-17-11293-2017, 2017.

Cong, Z., Kang, S., Kawamura, K., Liu, B., Wan, X., Wang, Z., Gao, S., and Fu, P.: Carbonaceous aerosols on the south edge of the Tibetan Plateau: concentrations, seasonality and sources, *Atmos. Chem. Phys.*, 15, 1573-1584, 10.5194/acp-15-1573-2015, 2015.

Bonasoni, P., Laj, P., Marinoni, A., Sprenger, M., Angelini, F., Arduini, J., Bonafè, U., Calzolari, F., Colombo, T., Decesari, S., Di Biagio, C., Di Sarra, A., Evangelisti, F., Duchi, R., Facchini, M. C., Fuzzi, S., Gobbi, G. P., Maione, M., Panday, A., Roccatò, F., Sellegri, K., Venzac, H., Verza, G., Villani, P., Vuillermoz, E., and Cristofanelli, P.: Atmospheric Brown Clouds in the Himalayas: first two years of continuous observations at the Nepal Climate Observatory-Pyramid (5079 m), *Atmospheric Chemistry and Physics*, 10, 7515-7531, 10.5194/acp-10-7515-2010, 2010.

Xu, J., Zhang, Q., Shi, J., Ge, X., Xie, C., Wang, J., Kang, S., Zhang, R., and Wang, Y.: Chemical characteristics of submicron particles at the central Tibetan Plateau: insights from aerosol mass spectrometry, *Atmos. Chem. Phys.*, 18, 427-443, 10.5194/acp-18-427-2018, 2018.

Junbo, W.: Daily meteorological Data of Nam Co Station China during 2019-2020, National Tibetan Plateau Data Center [dataset], 10.11888/Meteoro.tpdc.271782, 2021.

Venzac, H., Sellegri, K., Laj, P., Villani, P., Bonasoni, P., Marinoni, A., Cristofanelli, P., Calzolari, F., Fuzzi, S., Decesari, S., Facchini, M.-C., Vuillermoz, E., and Verza, G. P.: High frequency new particle formation in the Himalayas, *Proceedings of the National Academy of Sciences*, 105, 15666-15671, doi:10.1073/pnas.0801355105, 2008.

Shang, D., Hu, M., Zheng, J., Qin, Y., Du, Z., Li, M., Fang, J., Peng, J., Wu, Y., Lu, S., and Guo, S.: Particle number size distribution and new particle formation under the influence of biomass burning at a high altitude background site at Mt. Yulong (3410m), China, *Atmos. Chem. Phys.*, 18, 15687-15703, 10.5194/acp-18-15687-2018, 2018.

Due to harsh conditions and logistical limitations, our observation periods were limited. However, our conclusions are obvious and representative, and we will carry out more detailed observations for a longer period in the future if possible. To illustrate the representativeness of the observation periods, we have made supplements in the revised manuscript as follows:

## **“2.1 Measurement site**

The measurement was conducted from 26 April to 22 May, 2019 and 15 June to 25 June, 2019, and can be representative of the pre-monsoon season and the summer monsoon season, respectively (Text S1) (Bonasoni et al., 2010; Cong et al., 2015).”

## **“Text S1 The representativeness of the observation periods**

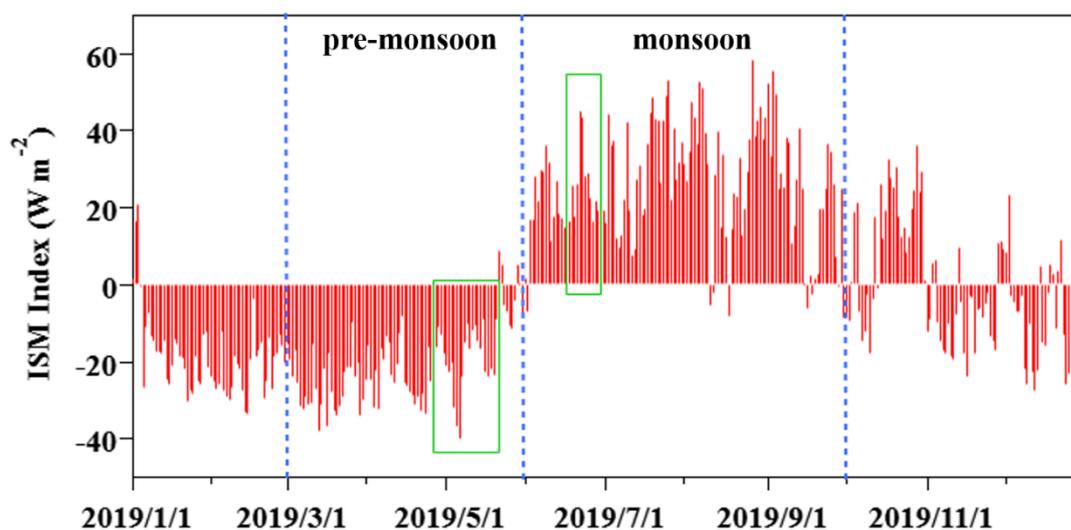
The measurement was conducted from 26 April to 22 May, 2019 and 15 June to 25 June, 2019, and can be representative of the pre-monsoon season and monsoon season, respectively.

Firstly, the intensity of Indian Summer Monsoon during the two measurements periods can represent that in

130 the whole pre-monsoon and monsoon seasons, respectively. The intensity of Indian Summer Monsoon is an  
131 important indicator to distinguish the monsoon season. Here the intensity of Indian Summer Monsoon (ISM) was  
132 indicated by the ISM Index, which are defined by the negative outgoing longwave radiation anomalies (with respect  
133 to the climatological annual cycle) averaged over the Bay of Bengal–India region (10°–25°N, 70°–100°E) (Wang  
134 and Fan, 1999). As shown in Fig. S1, the measurement periods (green boxes) were in the pre-monsoon season  
135 (March–May) and monsoon season (June–September), respectively. And the IMS index during the two  
136 measurements periods were equivalent to those of the whole pre-monsoon season (average: -19.5 vs -20.7 W m<sup>-2</sup>)  
137 and monsoon season (average: 27.0 vs 26.3 W m<sup>-2</sup>), respectively.

138 Secondary, the characteristics of meteorology and atmospheric pollutants in the two measurements periods  
139 was generally in agreement with the previous long-term studies at Nam Co station and other sites in the Tibetan  
140 Plateau (TP) (Yin et al., 2017; Cong et al., 2015; Bonasoni et al., 2010; Xu et al., 2018). That is, the characteristics  
141 of meteorology (temperature, WS and RH) and atmospheric pollutants (PM, BC and ozone) in the two  
142 measurements periods were matched with those in the whole pre-monsoon and monsoon season at Nam Co station  
143 and other sites in the TP.

144 Therefore, the two observation periods are representative in the seasonal characteristics in pre-monsoon season  
145 and monsoon season, respectively.



146  
147 **Figure S1.** The Indian Summer Monsoon (ISM) Index in 2019. The measurements periods are marked by the green boxes.”  
148

149 **2. Second, many of the conclusions made in the paper rely on SO<sub>2</sub> and VOC concentrations. Unfortunately,**  
150 **there are very limited measurements on these 2 trace gases (only VOCs during the monsoon season), instead**  
151 **their concentrations were estimated from large-scale model simulations. The simulated concentrations may**  
152 **have large uncertainties, which are not quantified by any means in the paper.**

153 Thanks for the comment. The model evaluation including the meteorological fields and air pollutants has

154 been added in the revised manuscript. In general, the results of model simulation of VOC and SO<sub>2</sub> show  
155 good performance in statistical parameters of model evaluation and correlation analysis with other tracers.  
156 The modelled VOC and SO<sub>2</sub> could be used for the NPF analysis. The detailed information can be found  
157 in the revised manuscript as follow:

### 158 “2.3 Model simulation

159 Considering the limited measurements on SO<sub>2</sub> and VOC in this observation (only VOC during pre-monsoon  
160 season), Weather Research and Forecasting/Community Multiscale Air Quality (WRF/CMAQ) modeling system  
161 was adopted to simulate the level of SO<sub>2</sub> and VOC in the whole observation period, to assist in the analysis of the  
162 role of sulfuric acid and organics in NPF events.

163 Weather Research and Forecasting (WRF) (version 4.2.1) model was used to simulate the meteorological  
164 conditions with the FNL reanalysis dataset. The 6 h FNL data were obtained from the U.S. National Centre for  
165 Atmospheric Research (NCAR), with a spatial resolution of 1.0° × 1.0° (<http://rda.ucar.edu/datasets/ds083.2/>, last  
166 accessed on 28 April 2022). The Community Multiscale Air Quality version 5.3.2 (CMAQv5.3.2) model, being one  
167 of the three-dimensional chemical transport models (CTMs) (Appel et al., 2021), configured with the gas-phase  
168 mechanism of SAPRC07tic and the aerosol module of AERO6i, was employed in this study to simulate the air  
169 quality over Tibet in the observation period (26 April to 22 May and 15 June to 25 June in 2019). Air quality  
170 simulations were performed with a horizontal resolution of 12 km. The corresponding domain covered Tibet and  
171 the surrounding countries and regions with 166 × 166 grids (Fig. S2), with the 18 layers in vertical resolution.  
172 Detailed information about the model setting is provided in Text S2.

173 The Multi-resolution Emission Inventory for China version 1.3 (MEICv1.3) (<http://www.meicmodel.org>) and  
174 Regional Emission inventory in ASia (REASv3.2) (<https://www.nies.go.jp/REAS/>) were used to provide the  
175 anthropogenic emissions from China and neighboring countries and regions, respectively. The MEICv1.3 emissions  
176 of the year 2019 were used. For REAS, the emission inventory in the year 2015 was used for 2019 as no emission  
177 inventory was released for the years after 2015. Although emission inventories are usually released 3 years behind,  
178 we acknowledge that this may cause additional uncertainties in the simulation. Biogenic emissions were generated  
179 using the Model for Emissions of Gases and Aerosols from Nature (MEGANv2.1) (Guenther et al., 2012). The open  
180 biomass burning emissions were processed using the Fire Inventory for NCAR (FINN) during the entire study  
181 period (Wiedinmyer et al., 2011).

182 The model evaluation is introduced in Text S2. The WRF and CMAQ models successfully reproduced the  
183 meteorological fields and air pollutants including PM and O<sub>3</sub> with model performance indices meeting the suggested  
184 benchmarks. For VOC, the observed VOC and predicted VOC in pre-monsoon season were compared to examine  
185 the model performance. The benchmarks for VOC had not been reported, but the statistical metrics of MFB (mean  
186 fractional bias, -0.47) and MFE (mean fractional error, 0.49) in this study are within the range reported in previous

VOC modelling result (Hu et al., 2017). The correlation coefficient (R) between simulated and observed VOC is 0.41, which reflected that the model can fairly simulate the variation of VOC concentration. It should be noted that VOC was underpredicted on the whole, which may due to the uncertainty of the emission inventory as mentioned before. For SO<sub>2</sub>, the WRF/CMAQ models have been successfully reproduced SO<sub>2</sub> in major regions in China with R of 0.25-0.79 (Mao et al., 2022). The simulated SO<sub>2</sub> level in the model domain is comparable with that measured at Mt. Yulong (Shang et al., 2018), with average values of 0.03±0.02 ppbv and 0.06±0.05 ppbv. At the same time, considering that both BC and SO<sub>2</sub> are mainly emitted from coal combustion and biomass burning, BC could be a good indicator for SO<sub>2</sub> especially for pristine environment without local anthropogenic source emissions. As shown in Fig.S6, a good correlation between SO<sub>2</sub> and BC measured at Mt. Yulong was found with correlation coefficient (R) of 0.79 (Shang et al., 2018). In this study, the modelled SO<sub>2</sub> and measured BC also showed good correlation with R of 0.58 (Fig. S6). In general, the results of model simulation showed good performance in statistical parameters and correlation analysis with other tracers. The modelled VOC and SO<sub>2</sub> may be helpful for the NPF analysis.”

## “2.3 Text S2 Model simulation

### Model Configurations

The meteorological conditions were simulated using the Weather Research and Forecasting (WRF) (version 4.2.1) model with the FNL reanalysis dataset. The 6 h FNL data were obtained from the U.S. National Centre for Atmospheric Research (NCAR), with a spatial resolution of 1.0° × 1.0° (<http://rda.ucar.edu/datasets/ds083.2/>, last accessed on 28 April 2022). The physical parameterizations used in this study are the Thompson microphysical process, RRTMG longwave/shortwave radiation scheme; Noah land-surface scheme; MYJ boundary layer scheme; and modified Tiedtke cumulus parameterization scheme. The detailed configuration settings could be found in the works of Hu et al. (2016), Mao et al. (2022), Wang et al. (2021a).

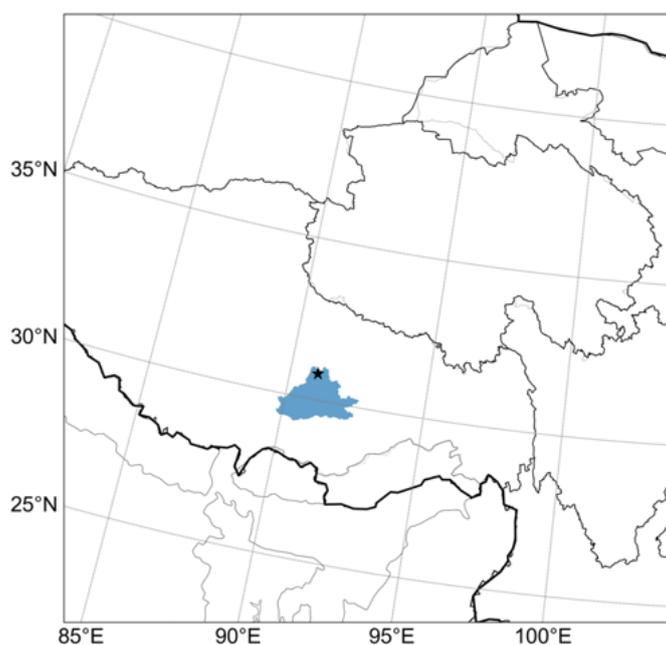
The Community Multiscale Air Quality version 5.3.2 (CMAQv5.3.2) model, being one of the three-dimensional chemical transport models (CTMs) (Appel et al., 2021), configured with the gas-phase mechanism of SAPRC07tic and the aerosol module of AERO6i, was employed in this study to simulate the air quality over Tibet from 24 April to 24 May and 13 June to 27 June in 2019, which contains the observation period. Air quality simulations were performed with a horizontal resolution of 12 km. The corresponding domain covered Tibet and the surrounding countries and regions with 166 × 166 grids (Fig. S2), with the 18 layers in vertical resolution. The initial and boundary conditions were provided by the default profiles. The simulated results of the first two days were not included in the model analysis, which served as a spin-up and reduced the effects of the initial conditions on the simulated results.

### Model Evaluation

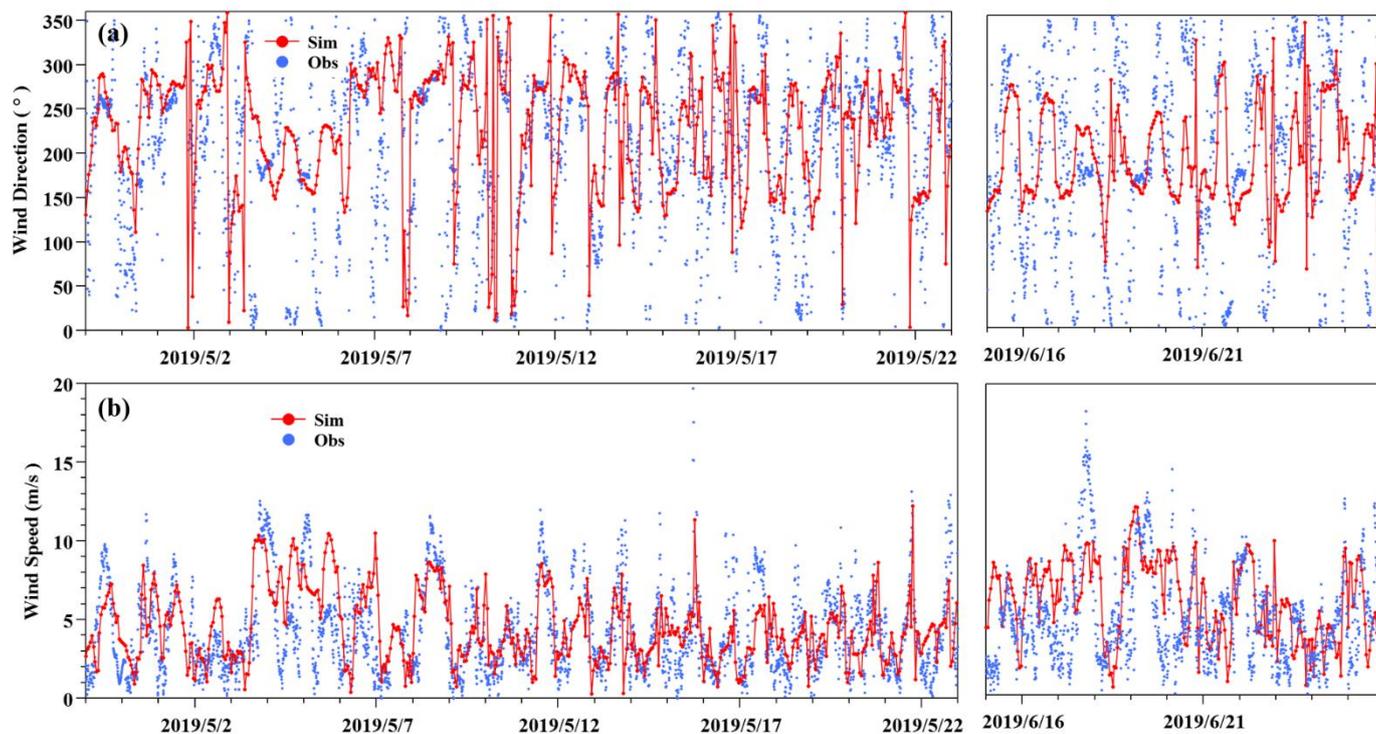
Previous studies have investigated the impacts of meteorological conditions on the formation, transportation,

220 and dissipation of air pollutants (Hu et al., 2016; Hua et al., 2021; Mao et al., 2022; Sulaymon et al., 2021b;  
221 Sulaymon et al., 2021a). Therefore, the evaluation of the WRF model performance was carried out before the usage  
222 of its meteorological fields in the CMAQ simulations. The evaluation of the WRF model was achieved by  
223 comparing the predicted wind speed (WS, m/s), wind direction (WD, °) at 10 m above the surface, RH (%) and  
224 temperature (T, °C) to the observed values. Fig. S3 showed that WS was well simulated both in pre-monsoon and  
225 monsoon seasons. WD was well simulated in pre-monsoon season, and there seems to be some deviation in the  
226 simulation of north wind in monsoon season. The main reason about the deviation in WD may be due to the poor  
227 terrain and complicated weather conditions. Nevertheless, both simulations and measurements showed more  
228 frequent southerly winds during monsoon season. RH and temperature were well simulated in the whole periods  
229 (Fig. S4). The good model performance with the statistical metrics of WS, RH and temperature meeting the  
230 suggested benchmarks are shown in Table S1. Generally, the simulated meteorological fields were qualified and can  
231 be further utilized in driving the CMAQ model

232 Fig. S5 showed the comparison of simulated hourly mean concentration about PM, O<sub>3</sub> and VOC in observation  
233 site, which were simulated by CMAQ. The statistical indices used in evaluating the CMAQ model were present in  
234 Table S2. It can be seen that PM and O<sub>3</sub> meet the suggested benchmarks, which reflect the good model performance.  
235 The observed VOC and predicted VOC in pre-monsoon season were compared to examine the model performance.  
236 The benchmarks for VOC had not been reported, but the MFB (mean fractional bias) and MFE (mean fractional  
237 error) values are within the range reported in previous VOC modelling result (Hu et al., 2017). The correlation  
238 coefficient (R) between simulated and observed VOC is 0.41, which reflected that the model can fairly simulate the  
239 variation of VOC concentration. It should be noted that VOC was underpredicted on the whole, which may due to  
240 the uncertainty of the emission inventory.



241  
242 **Figure S2.** WRF/CMAQ modeling domain



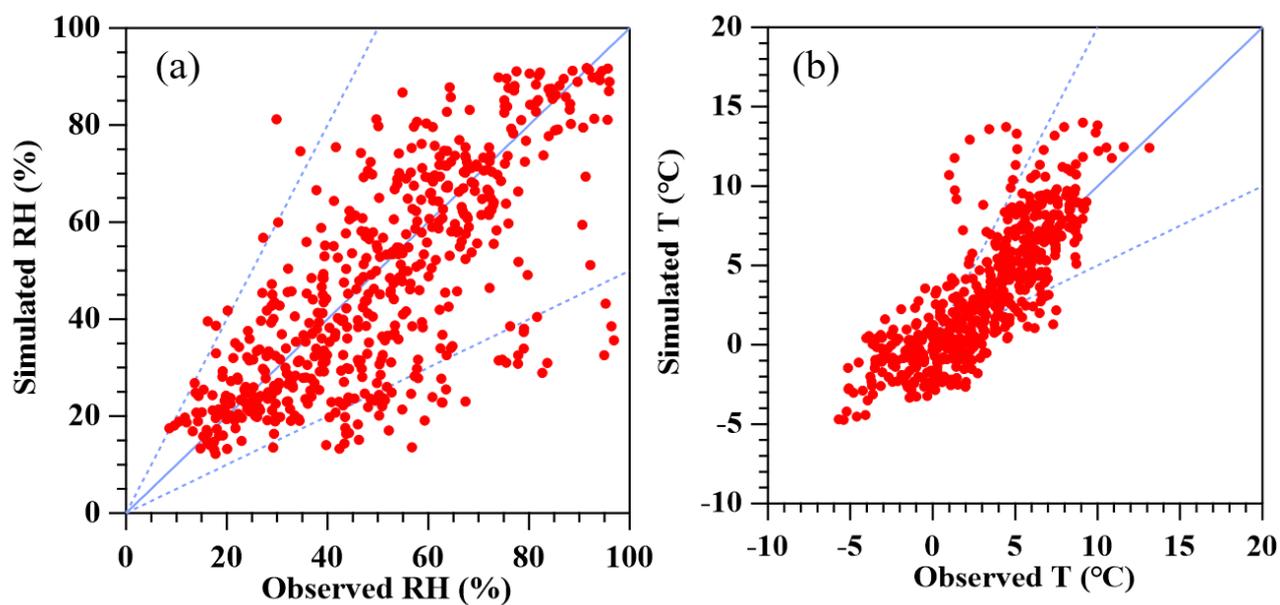
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**Figure S3.** Comparison of simulated (in red dot-line) and observed (in blue dot) wind direction (WD, °) and wind speed (WS, m/s). Observed is 10 minutes mean data. Simulated is hourly mean data.

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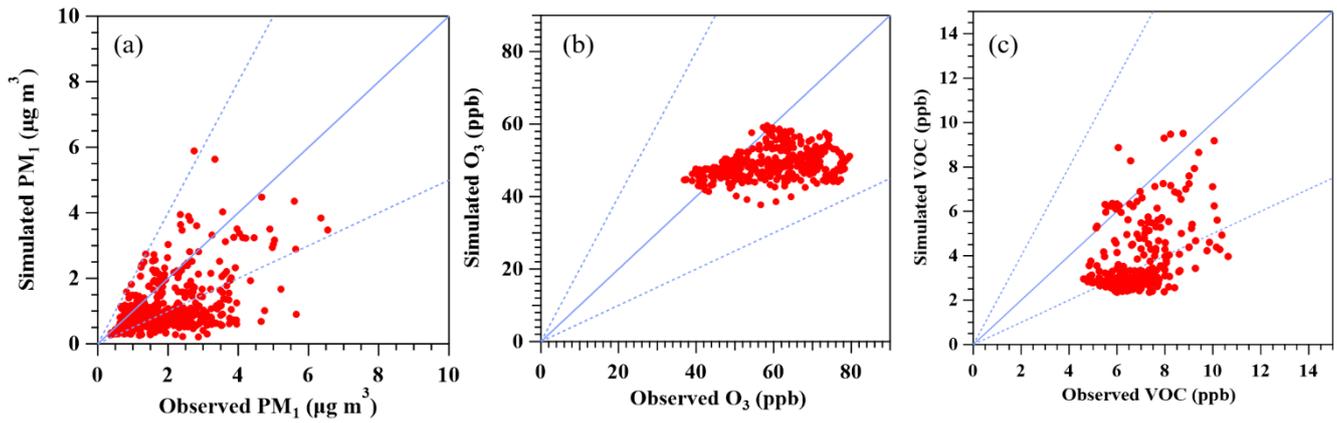
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**Figure S4.** Comparison of simulated and observed RH (%) and temperature (T, °C). RH and temperature are hourly mean data.

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**Figure S5.** Comparison of simulated and observed PM ( $\mu\text{g}/\text{m}^3$ ),  $\text{O}_3$  (ppb) and VOC (ppb). PM,  $\text{O}_3$  and VOC are hourly mean concentration.

**Table S1.** Model performance of meteorological factors at Nam Co station

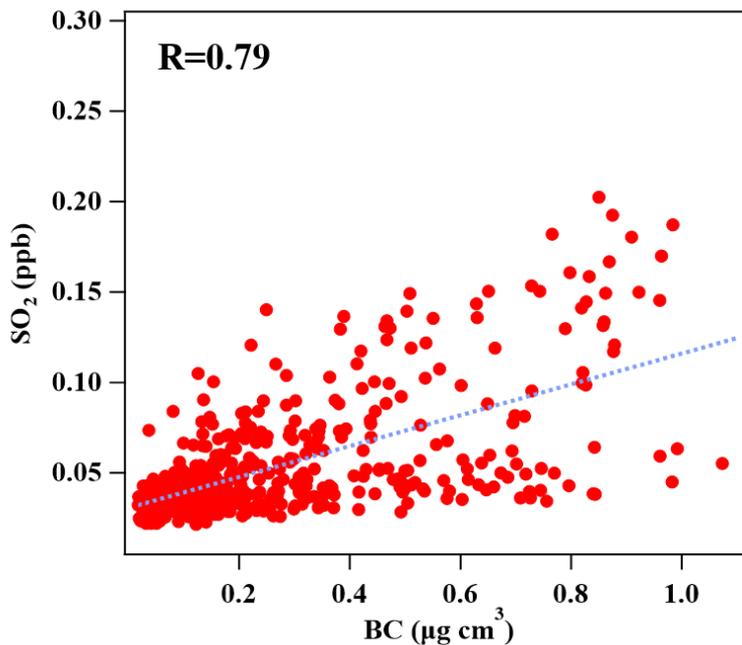
	WS				RH				T			
	MB	ME	RMSE	R	MB	ME	RMSE	R	MB	ME	RMSE	R
<b>Statistic</b>	0.42	0.87	1.20	0.51	-1.38	12.20	16.30	0.67	0.07	1.85	2.43	0.89
<b>Benchmarks</b>	$\leq \pm 0.5$	$\leq 2.0$	$\leq 2.0$						$\leq \pm 0.5$	$\leq 2.0$		

*MB: mean bias; ME: mean error; RMSE: root mean square error; R: correlation coefficient. The benchmarks were suggested by Boylan and Russell (2006).*

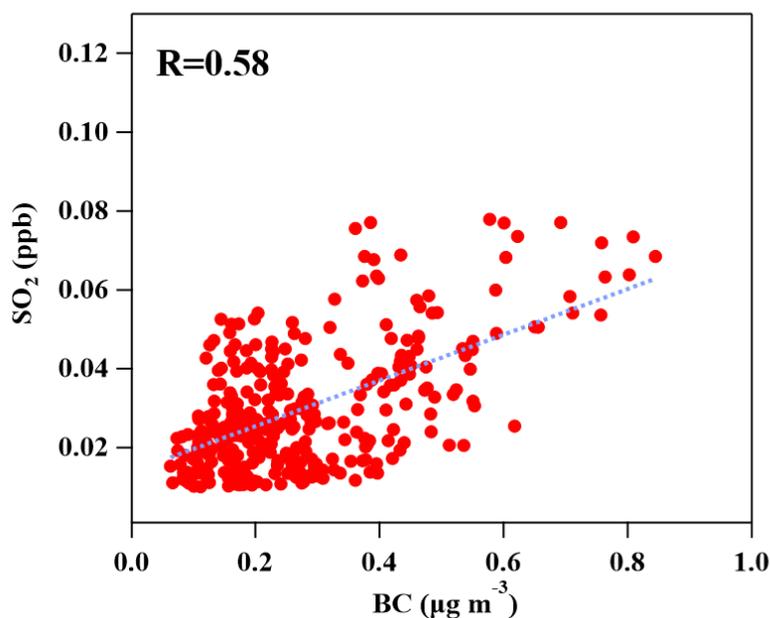
**Table S2.** Model performance of the air pollutants at Nam Co station

	PM <sub>1</sub>			O <sub>3</sub>			VOC			SO <sub>2</sub>		
	MFB	MFE	R	NMB	NME	R	MFB	MFE	R	NMB	NME	R
<b>Statistic</b>	0.49	0.50	0.72	0.14	0.23	0.51	-0.47	0.49	0.41			
<b>Benchmarks</b>	$< \pm 0.6$	$< 0.75$	$> 0.4$	$< \pm 0.15$	$< 0.35$	$> 0.5$						
<b>References</b>							$< \pm 0.77$	$< 0.74$		$< \pm 4.38$	$< \pm 4.38$	0.25- 0.79

*NMB: normalized mean bias; NME: normalized mean error; R: correlation coefficient; MFB: mean fractional bias; MFE: mean fractional error. The benchmarks for PM and O<sub>3</sub> were suggested by Emery et al. (2017) and Boylan and Russell (2006), respectively. The references for VOC and SO<sub>2</sub> were from Hu et al. (2017) and Mao et al. (2022), respectively.*



266  
267 **Figure S6.** Relationship between SO<sub>2</sub> and BC at Mt. Yulong in 2015. The correlation coefficient R is 0.79.



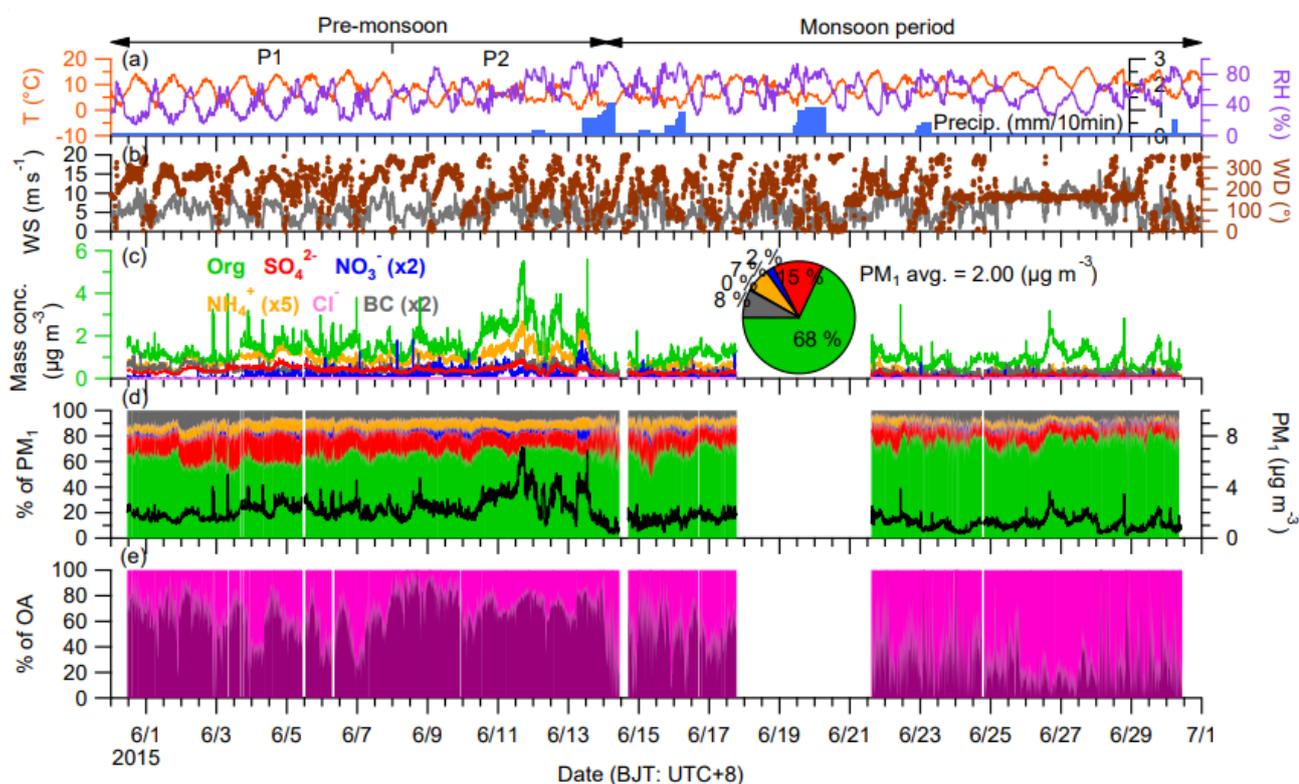
268  
269 **Figure S7.** Relationship between modelled SO<sub>2</sub> and BC at Nam Co station. The correlation coefficient R is 0.58.’

270  
271 **Reply to Reviewer 1’s important scientific comments (5):**

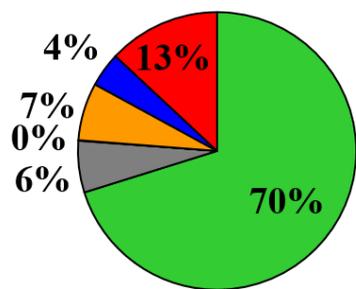
272 *1. The description on how CCN concentrations were calculated (section 2.4) is incomplete. Apparently, the*  
 273 *authors used equations 4 and 5 to determine the critical diameters corresponding to different*  
 274 *supersaturations, and from these critical diameters one then gets the number of CCN using measured*  
 275 *particle number size distributions. However, this calculation cannot be done without knowing the*  
 276 *hygroscopicity parameter cappa. Did the authors simply assumed a fixed value for cappa or did they*  
 277 *estimate it from some chemical data?*

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Thanks for the comment. The hygroscopicity parameter kappa was assumed to be a constant value of 0.12 throughout the measurement period, according to the previous measurement at Mt. Yulong on the southeastern TP (Shang et al., 2018). This is mainly due to the similar proportions of chemical components between Nam Co station (Xu et al., 2018) and Mt. Yulong with around 70% fraction of organics (Fig R5). And the fractions of chemical components at Nam Co station were stable.



283



284

285 **Figure R5.** The combo plot of the data of the Nam Co study including (a) the meteorological conditions (T :  
286 air temperature; RH: relative humidity; Precip.: precipitation), (b) the variation of WS (wind speed) colored  
287 according to WD (wind direction), (c) the temporal variation of mass concentration of PM<sub>1</sub> species and the  
288 average contribution of each species (pie chart), (d) the mass contribution of each PM<sub>1</sub> species and the total  
289 mass concentration of PM<sub>1</sub>, and (e) the mass contribution of PMF results. Three periods based on the  
290 meteorological conditions were marked (Xu et al., 2018) (top). The average contribution of each chemical  
291 species at Mt. Yulong on the southeastern TP (Shang et al., 2018) (bottom).

292

293

In addition, the kappa at Nam Co station in pre-monsoon and monsoon seasons can be estimated by using

the previously measured chemical data at this site (Xu et al., 2018). The kappa of the mixed particles was calculated based on  $\kappa$ -Köhler theory and the Zdanovskii–Stokes–Robinson (ZSR) mixing rule (Petters and Kreidenweis, 2007). The values of  $\kappa$  is 0.48 for  $(\text{NH}_4)_2\text{SO}_4$  and 0.58 for  $\text{NH}_4\text{NO}_3$  (Petters and Kreidenweis, 2007). The  $\kappa$  is assumed to be 0.1 for organics (Wu et al., 2015). We derived the volume fraction of each species by dividing mass concentration by its density. The densities are  $1.77 \text{ g cm}^{-3}$  for  $(\text{NH}_4)_2\text{SO}_4$  and  $1.72 \text{ g cm}^{-3}$  for  $\text{NH}_4\text{NO}_3$ . The densities of organics are assumed to be  $1.2 \text{ g cm}^{-3}$  (Fan et al., 2020). The  $\kappa$  and density of BC are assumed to be 0 and  $1.7 \text{ g cm}^{-3}$ . It was found that average value of kappa was 0.15 and 0.13 in pre-monsoon and monsoon seasons, respectively. The  $D_c$  at  $S_c$  levels of 0.6% and 1.2% were  $72.4 \pm 1.0$  and  $45.7 \pm 0.6$  nm in pre-monsoon season, and  $69.1 \pm 0.9$  and  $43.6 \pm 0.6$  nm in monsoon season. It was comparable with that used in this study ( $\kappa$ : 0.12,  $S_c=0.6\%$ :  $73.4 \pm 1.3$ ,  $S_c=1.2\%$ :  $46.3 \pm 0.8$  nm). And it had little effect on the final result of CCN concentration. Therefore, we finally decided to adopt the fixed  $\kappa$  value of 0.12.

Shang, D., Hu, M., Zheng, J., Qin, Y., Du, Z., Li, M., Fang, J., Peng, J., Wu, Y., Lu, S., and Guo, S.: Particle number size distribution and new particle formation under the influence of biomass burning at a high altitude background site at Mt. Yulong (3410&thinsp;m), China, Atmos. Chem. Phys., 18, 15687-15703, 10.5194/acp-18-15687-2018, 2018.

Xu, J., Zhang, Q., Shi, J., Ge, X., Xie, C., Wang, J., Kang, S., Zhang, R., and Wang, Y.: Chemical characteristics of submicron particles at the central Tibetan Plateau: insights from aerosol mass spectrometry, Atmos. Chem. Phys., 18, 427-443, 10.5194/acp-18-427-2018, 2018.

Wu, Z. J., Poulain, L., Birmili, W., Größ, J., Niedermeier, N., Wang, Z. B., Herrmann, H., and Wiedensohler, A.: Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of hygroscopicity measurements, Atmos. Chem. Phys., 15, 13071-13083, 10.5194/acp-15-13071-2015, 2015.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961-1971, 10.5194/acp-7-1961-2007, 2007.

Fan, X., Liu, J., Zhang, F., Chen, L., Collins, D., Xu, W., Jin, X., Ren, J., Wang, Y., Wu, H., Li, S., Sun, Y., and Li, Z.: Contrasting size-resolved hygroscopicity of fine particles derived by HTDMA and HR-ToF-AMS measurements between summer and winter in Beijing: the impacts of aerosol aging and local emissions, Atmos. Chem. Phys., 20, 915-929, 10.5194/acp-20-915-2020, 2020.

### “3.4 Significant increase of CCN in monsoon season

The CCN concentration was estimated following the method introduced in Sect. 2.4. Considering the similar proportions of chemical components between Nam Co station and Mt. Yulong with around 70% fraction of organics, and the stability of the fractions of chemical components, the hygroscopicity parameter  $\kappa$  was assumed to be a constant value of 0.12 throughout the measurement period according to the previous measurement at Mt. Yulong in the TP (Shang et al., 2018).”

**2. The discussion on the role of condensation sink (CS) in favoring/disfavoring NPF is not logical. The authors first say that their result differs from those found in earlier studies (lines 243-244), but then mention a few studies which actually agree with their findings (lines 245-247). Please reformulate this part**

331 *of the text, as it causes confusion in its present form.*

332 Thanks for the comment. Considering the possible confusion, we have reformulated the discussion in the  
333 revised manuscript as follows:

### 334 “3.3.1 Condensation sink

335 CS is the key factor controlling the occurrence of NPF events especially in urban environment (Yan et al.,  
336 2021). At some high-altitude observations at a larger scale, the important role of the transported pre-existing  
337 particles in NPF events was also emphasized (Rose et al., 2019; Boulon et al., 2010). Here we analyzed the CS  
338 levels in NPF days and non-event days at Nam Co station. As shown in Fig. 3a, the levels of CS in NPF-pre days,  
339 NPF-monsoon days and non-event days were approximate during 11:00–18:00 (the occurrence time of NPF events),  
340 although the CS in the early morning of NPF-pre days seems to be slightly lower. The CS was mainly in the range  
341 of  $0.1 \times 10^{-2}$ – $0.15 \times 10^{-2} \text{ s}^{-1}$  during the NPF occurrence time, which was much lower than that at most locations in  
342 China, such as  $\sim 0.01 \text{ s}^{-1}$  in urban Beijing (Deng et al., 2021), and  $0.1 \times 10^{-2}$  to  $28.4 \times 10^{-2} \text{ s}^{-1}$  at Mt. Tai (Lv et al.,  
343 2018), and comparable with that at Mt. Yulong ( $\sim 0.2 \times 10^{-2} \text{ s}^{-1}$ ) (Shang et al., 2018). The result varied from previous  
344 studies which reported much lower CS during NPF days than that in non-event days (Zhou et al., 2021; Lv et al.,  
345 2018). It indicated that CS was not the decisive factor controlling the occurrence of NPF events at Nam Co station.”

346  
347 *3. The discussion on the role of VOCs (lines 266-277) could be improved as well. First, considering the*  
348 *typical variability of VOC concentrations in ambient measurements, I would think a 20% higher VOC*  
349 *concentration is slightly rather than noticeably higher (line 268). It is also confusing that for the pre-*  
350 *monsoon season the VOC concentration difference is given as % while for the monsoon season it is given*  
351 *as an absolute value (ppb).*

352 Thanks for the comment. This part has been modified in the revised manuscript as follows:

### 353 “3.3.1 Gas precursors

354 In addition to sulfuric acid, organics are also considered to be an important factor of NPF events. Observations  
355 and laboratory experiments have found that organics may participate in or even dominate the nucleation and growth  
356 process in NPF events in pristine environments and the preindustrial atmosphere. For example, CLOUD (Cosmics  
357 Leaving Outdoor Droplets) experiments observed obvious NPF events from highly oxidized organics without the  
358 involvement of sulfuric acid (Kirkby et al., 2016). At the high-altitude sites of Jungfrauoch and Himalaya, NPF  
359 events occurred mainly through the condensation of highly oxygenated molecules (HOMs) (Bianchi et al., 2016;  
360 Bianchi et al., 2021). Due to instrument status, VOC measurement was only available in pre-monsoon season. The  
361 concentration of VOC (total VOC) showed a higher value (20%) during 11:00-18:00 on NPF-pre days compared  
362 with non-event days (Fig. 3c). Aromatics, which can be used as the indicator of anthropogenic emissions, also  
363 exhibited a higher level (20%) during NPF-pre days (Fig. 3d). This suggested that VOC may be the key factor in

364 determining the occurrence of NPF events. In order to further evaluate the role of VOC, we used WRF/CMAQ  
365 models to simulate the spatial distribution of VOC concentration in pre-monsoon and monsoon seasons. The  
366 detailed information about the model setting and evaluation can be found in Text S2. As shown in Fig.4, the  
367 simulated VOC levels in NPF days including NPF-pre and NPF-monsoon days were higher than those in non-event  
368 days. The average modelled VOC concentrations in NPF-pre days and NPF-monsoon days were 25% and 88%  
369 higher than those in non-event days, respectively. Therefore, we further considered that the organic matter could be  
370 the key factor to determine whether NPF event occurred. Nucleation of pure organics or organics involved could be  
371 the dominant mechanism at Nam Co station. In addition, higher organic concentrations were observed in monsoon  
372 season (NPF-monsoon days) compared with those in pre-monsoon season (NPF-pre days and non-event days). The  
373 result is consistent with one recent research which has found that the concentration of monoterpene-derived HOMs  
374 in East Asia was higher in summer (June-August) than that in Spring (March-May) by using GEOS-Chem global  
375 chemical transport model (Xu et al., 2022). This means that the frequent NPF events in monsoon season could be  
376 caused by the higher organic matters.

377  
378 ***4. Wind direction is a very local quantity, and does not necessary tell correctly air pollutant sources or***  
379 ***transport pathways. I wonder whether the authors have information on air mass trajectories which would***  
380 ***provide more direct support for their statements on lines 283-295.***

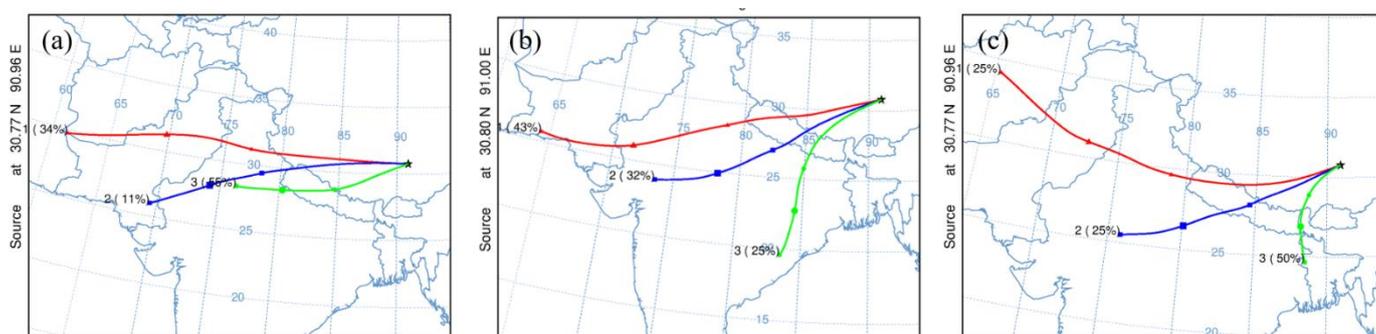
381 Thanks for the comment. The air mass trajectories have been added in the revised manuscript as follpws:

#### 382 “3.3.1 Air mass origins and meteorology

383 While the concentrations of organic precursors could be the most possible reason for the occurrence of NPF  
384 events, the external factors driving the difference in VOC levels between the NPF and non-event days and other  
385 conditions that may affect the characteristics of NPF were still unknown. This indicated that a further investigation  
386 into other NPF-related variables was still required.

387 There are almost no local anthropogenic source emissions at Nam Co station. The air pollutants at the  
388 observation site mainly brought by air mass transmission. Backward trajectories were utilized to identify the air  
389 mass origins associated with NPF events. The frequencies of the 48 h back trajectories of air masses arriving at  
390 Nam Co station during the occurrence time of nucleation (11:00-18:00) in non-event days, NPF-pre days and NPF-  
391 monsoon days were present in Fig. 5. It can be found that the dominant air masses in non-event days were from the  
392 west (almost 100%) and passed by western Nepal, northwest India and Pakistan. In comparison, air masses in NPF-  
393 pre days and NPF-monsoon days mainly come from the south and southwest (57% and 75%) and originated in the  
394 northeast India. WD, which can reflect the local situation for air masses and the source of air pollutants, showed a  
395 high frequency of strong southerly wind in NPF-pre days and NPF-monsoon days and westerly wind in non-event  
396 days (Fig. S14). The example of the spatial distribution of wind field in non-event days, NPF-pre days and NPF-

monsoon days displayed the similar phenomenon on a larger scale (Fig. S15). These results suggested that the occurrence of NPF events was related to the southerly and southwesterly air masses. When the southerly air mass occurred, it may bring organic precursors from the southern region (northeast India) to Nam Co station, driving the occurrence of NPF events in this area. As for the significantly higher NPF frequency in monsoon season, it resulted from the more frequent southerly air masses (summer monsoon) in monsoon season in comparison with pre-monsoon season as introduced in Sec. 3.1. The summer monsoon can bring the higher organic concentrations in monsoon season (NPF-monsoon days) compared with those in pre-monsoon season (NPF-pre and non-event days) (Fig.4), thus triggered almost daily NPF events. Similar result was found in the recent study which showed that the Indian summer monsoon acted as a facilitator for transporting gaseous pollutants (Yin et al., 2021).



**Figure 4.** The frequencies of the 48 h back trajectories of air masses arriving at Nam Co station from different directions during the occurrence time of nucleation (11:00-18:00) in (a) non-event days, (b) NPF-pre days and (c) NPF-monsoon days.”

**5. The enhancements of CCN concentrations due to NPF is reported in 3 different ways in section 3.4: 1) using an enhancement factor EF, 2) using percentage increases, and 3) stating that something is N times higher than... This is confusing. I highly recommend the authors to unify this discussion.**

Thanks for the comment. Considering the possible confusion, we have unified the discussion (N times higher than) in the revised manuscript as follows:

### “3.4 Significant increase of CCN in monsoon season

The high-frequency NPF events in the summer monsoon period markedly increased the number concentration of atmospheric aerosols. The average PNSD during pre-monsoon and monsoon seasons were plotted in Fig. 7a with much higher number concentrations observed during monsoon season. The mean total particle number concentration (PN<sub>4-700</sub>) in monsoon season was  $3647 \pm 2671 \text{ cm}^{-3}$ , which was more than 2 times higher than that of pre-monsoon season ( $1163 \pm 1026 \text{ cm}^{-3}$ ). Although the measured particle size ranges were not the same with other studies, the results can still be comparable as the background particles were mainly distributed in tens to hundreds of nanometers. As shown in Table 1, PN<sub>4-700</sub> at Nam Co station in pre-monsoon season was comparable with other high-altitude sites around the world, while PN<sub>4-700</sub> in monsoon season was much higher due to the frequent NPF events. The atmospheric particles contributed by new particle nucleation and growth in monsoon season were mainly concentrated below 100 nm. Among them, the

concentration of Nucleation-mode particles in monsoon season was about 2 times higher than that in pre-monsoon season, and the concentration of Aitken-mode particles was 3.5 times higher than that in pre-monsoon season. In contrast, Accumulation-mode particles (>100 nm) which were related to the secondary formation process and long-range transport (Wang et al., 2013; Vu et al., 2015), were nearly at the same level in the two seasons (around 200 cm<sup>-3</sup>). As for CCN, the average number concentrations of CCN at  $S_c$  of 0.6% and 1.2% in monsoon season were 434±242 and 863±628 cm<sup>-3</sup>, respectively (Fig. 8). The results were about 0.1 and 0.6 times higher than those in pre-monsoon season at  $S_c$  of 0.6% (396±177 cm<sup>-3</sup>) and 1.2% (552±261 cm<sup>-3</sup>).

In addition to the average particle number concentration in the two seasons, the important impact of NPF events is more reflected in the increased number concentration of aerosol and CCN in a short time, that is, the aerosol/ CCN production. The aerosol production and CCN production during an NPF event can be obtained by comparing the particle number concentration at the beginning of the increase of the target particles ( $N_{init}$ ) with the maximum number concentration ( $N_{max}$ ), as introduced by Rose et al. (2017). The  $N_{init}$  and  $N_{max}$  are hourly average concentrations as shown in Fig. S16. Fig. 9 showed the average daily variation of PNSD, three modes particles and CCN in the whole periods in pre-monsoon season and monsoon season. In monsoon season, Nucleation-mode particles (4-25 nm) started to increase quickly at around 11:00 when nucleation occurred. The freshly nucleated particles grew to larger sizes due to condensation and coagulation of the pre-existing particles within several hours, which contributed to the increase of Aitken-mode particles (25-100 nm) from 15:00. The average daily aerosol production of Nucleation-mode particles and Aitken-mode particles in monsoon season was around 3400 cm<sup>-3</sup> and 1200 cm<sup>-3</sup>, respectively. As for CCN, the average production of CCN at  $S_c$  of 0.6% and 1.2% in monsoon season was 180 and 518 cm<sup>-3</sup>, respectively. The production of CCN were lower than previous studies because they only considered NPF days and their environment was relatively polluted (Rose et al., 2017; Shen et al., 2016). The production of aerosols and CCN was much lower in pre-monsoon season, although the particles and CCN at around 11:00 were comparable with monsoon season. The average daily aerosol production of Nucleation-mode particles and Aitken-mode particles in pre-monsoon season were around 500 cm<sup>-3</sup> and 300 cm<sup>-3</sup>, respectively. And the CCN production at  $S_c$  of 0.6% and 1.2% was 160 and 286 cm<sup>-3</sup>, respectively. The average daily production of Nucleation-mode particles, Aitken-mode particles and CCN at  $S_c$  of 1.2% in monsoon season was 5.8, 3 and 0.8 times higher than that in pre-monsoon season, respectively."

### Reply to Reviewer 1's minor comments (5):

*1. line 49: I suppose that the authors mean quantities like the particle formation and growth rate as referring to parameters of NPF. I do not feel that parameter is a good wording here, rather suggesting something as characteristics of NPF.*

Thanks for the help comment. We have made correction in the revised manuscript.

457 **2. lines 127-128: Classification of a NPF event seems untypical. Has the performance of this classification**  
458 **method tested and has it been used in other studies besides Fang et al. (2020)? The word obviously does**  
459 **not fit into this context.**

460 Thanks for the comment. The classification method of NPF events has been used in Tang et al (2021) and  
461 Shang et al (2022). Considering the possible misunderstanding, we have described the classification  
462 method in more detail in the revised manuscript.

463 Tang, L., Shang, D., Fang, X., Wu, Z., Qiu, Y., Chen, S., Li, X., Zeng, L., Guo, S., and Hu, M.: More Significant Impacts From  
464 New Particle Formation on Haze Formation During COVID-19 Lockdown, *Geophysical Research Letters*, 48,  
465 e2020GL091591, <https://doi.org/10.1029/2020GL091591>, 2021.

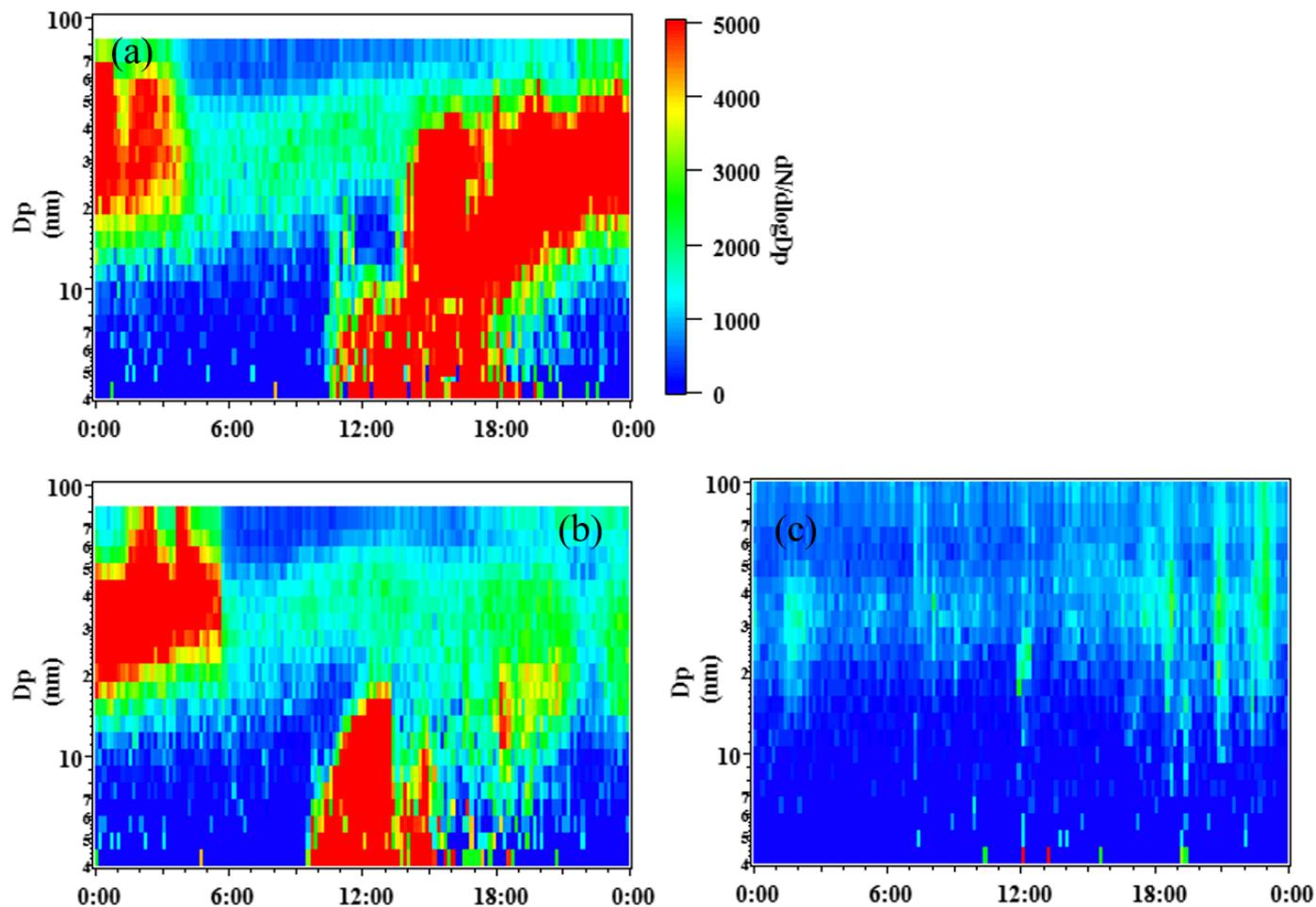
466 Shang, D., Tang, L., Fang, X., Wang, L., Yang, S., Wu, Z., Chen, S., Li, X., Zeng, L., Guo, S., and Hu, M.: Variations in source  
467 contributions of particle number concentration under long-term emission control in winter of urban Beijing,  
468 *Environmental Pollution*, 304, 119072, <https://doi.org/10.1016/j.envpol.2022.119072>, 2022.

### 469 **“2.3 Parameterization of NPF**

470 In this study, a typical NPF event was defined by that there is a burst in the 3-10 nm particles number concentration  
471 ( $PN_{3-10}$ ) and subsequent growth of these newly formed particles (Fang et al., 2020; Dal Maso et al., 2005). The days  
472 without newly particle formation were defined as non-event days. Days in which the increase of  $PN_{3-10}$  was observed  
473 without the particles growing to the larger size, or days when we can see the later phase of a mode growing in the Aitken  
474 mode size range were treated as undefined days (Dal Maso et al., 2005). The examples of the classification of NPF events  
475 are shown in Fig.S8.

476 Fang, X., Hu, M., Shang, D., Tang, R., Shi, L., Olenius, T., Wang, Y., Wang, H., Zhang, Z., Chen, S., Yu, X., Zhu,  
477 W., Lou, S., Ma, Y., Li, X., Zeng, L., Wu, Z., Zheng, J., and Guo, S.: Observational Evidence for the  
478 Involvement of Dicarboxylic Acids in Particle Nucleation, *Environmental Science & Technology Letters*, 7,  
479 388-394, [10.1021/acs.estlett.0c00270](https://doi.org/10.1021/acs.estlett.0c00270), 2020.

480 Dal Maso, M., Kulmala, M., Riipinen, I., and Wagner, R.: Formation and growth of fresh atmospheric aerosols:  
481 Eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, *Boreal Environment Research*,  
482 10, 323-336, 2005.



483 **Figure S8.** Typical particle number size distributions of (a) a NPF day (23 June, 2019), (b) an undefined day (24 June, 2019),  
 484 and (c) a non-event day (15 May, 2019).”  
 485  
 486

487 **3. lines 225-233: The authors list a number of things that potentially affect the occurrence of NPF. The list**  
 488 **misses one highly relevant quantity: the intensity of solar radiation. This quantity should be mentioned**  
 489 **here.**

490 Thanks for the comment. We have made correction in the revised manuscript as follows:

491 “Whether an NPF event can occur is mainly related to 1) the CS, which mainly referred to the scavenging rate of  
 492 precursors, clusters, and newly formed particles by background aerosols. High CS can lead to the continual reduction in  
 493 newly formed particle number concentration, and inhibit the occurrence of NPF; 2) the gaseous precursors that can  
 494 participate in nucleation and growth, including sulfuric acid (Kulmala et al., 2013), dimethylamine (Yao et al., 2018),  
 495 ammonia (Xiao et al., 2015) and VOC (Tröstl et al., 2016; Fang et al., 2020; Qiao et al., 2021). A sufficiently high  
 496 concentration of low volatility vapors (precursors) can contribute to persistent nucleation and generating new  
 497 atmospheric particles; 3) air mass origins and meteorological factors including WD, RH, temperature, the intensity of  
 498 solar radiation, etc, which can influence the occurrence and intensity of NPF events by directly or indirectly affecting  
 499 the source and sink parameters.”

502 **4. Unlike the Aitken mode, the nucleation mode is usually written using a lower-case letter (lines 326, 359,**  
503 **383)**

504 Thanks for the comment. The nucleation mode is written using a lower-case letter in the revised  
505 manuscript:

506 “nucleation mode particles”

507  
508 **5. In several places (lines 225, 254, 258, 261, 284, 296, 299, 340), the use of tense is somewhat wrong, or at**  
509 **least uncommon. Please reconsider which tense to use in these places.**

510 Thanks for the comment. We have made correction in the revised manuscript as follows:

511 “Whether an NPF event can occur is mainly related to 1) the CS, which mainly referred to the scavenging rate of  
512 precursors, clusters, and newly formed particles by background aerosols.”

513 “Gaseous sulfuric acid is identified as the key precursor for nucleation and initial growth due to its low  
514 volatility (Kulmala et al., 2013; Qiao et al., 2021).”

515 “In addition to sulfuric acid, organics are also considered to be an important factor of NPF events. Observations  
516 and laboratory experiments have found that organics may participate in or even dominate the nucleation and growth  
517 process in NPF events in pristine environments and the preindustrial atmosphere.”

518 “The frequencies of the 48 h back trajectories of air masses arriving at Nam Co station during the occurrence time  
519 of nucleation (11:00-18:00) in non-event days, NPF-pre days and NPF-monsoon days are present in Fig. 5.”

520 “In Fig. 6, we show the diurnal variations of meteorological factors during NPF-pre days, NPF-monsoon days and  
521 non-event days at Nam Co station.”

522 “The similar temperature in NPF-pre days and non-event days suggests that temperature is not a crucial factor for  
523 NPF event occurrence.”

524 “The average PNSD during pre-monsoon and monsoon seasons are plotted in Fig. 7a with much higher number  
525 concentrations observed during monsoon season.”

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