Reply to comments

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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 revised version of the manuscript is greatly improved compared with the original one. There are, however, some minor issues that need to be considered before I can recommend acceptance of the paper for publication.

We appreciate the comments from the reviewer on this manuscript. We have answered them 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 important comments (5):

1. Concerning the calculation of CCN concentrations, I suggest reformulating as "…was assumed to be equal to 0.12 throughout the …" (lines 356-357). Furthermore, the statement on lines 360-361 is scientifically incorrect: a paper published in 2018 cannot say anything on measurements conducted in 2019 (the data of this paper). Please reformulate.

Thanks for the comment. We have reformulated the expressions in the revised manuscript as follows:

"The hygroscopicity parameter κ was assumed to be equal to a constant value of 0.12 throughout the measurement period according to the previous measurement at Mt. Yulong in the TP."

"There could be uncertainties in the values of κ due to the variation of chemical components, but they had little impact on D_c thus the final result of CCN concentration."

2. Concerning the tense, one should write this suggests/indicates, not suggested/indicated (several places

in the text), … are shown … (line 195), … is comparable with … (line 206), … and represents (line 207), … is comparable with (lines 230, 237 and 240), … results are most … (line 349), Fig. 9 shows … (line 382)

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

3. When discussing the different seasons, one should add "the" (e.g. in the monsoon seasons etc.). Also, I would write "the Nam Co station".

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

4. To make the text more readable, the numerical values of CS could be written in the form 0.02 etc rather than using an exponential form (several places in the text).

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

5. Lines 23 and 413: I would recommend reformulating: …extreme/evident seasonal differences, with 15% and… Also, decide whether this difference is extreme or evident, as these two are quite different characteristics.

Thanks for the comment. We have reformulated the expressions in the revised manuscript as follows:

"The frequencies of NPF events exhibited **evident seasonal differences** with 15% in the pre-monsoon season and 80% in the monsoon season."

"The most important finding of this study was that there were **evident seasonal differences** in the frequencies of NPF events at the Nam Co station with 15% in the pre-monsoon season and 80% in the monsoon season."

Line 60: … but not to biogenic…

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

"At Mt. Yulong on the southeastern TP, the NPF frequency was only 14% during the pre-monsoon season and the occurrence of NPF events was related to an elevated boundary layer or transported biomass burning pollutants from southern Asia, **but not to biogenic** condensable vapours (Shang et al., 2018; Du et al., 2015)."

Line 63: … may be associated with …

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

"These results indicated that the frequency and mechanism of NPF **may be associated with** air mass origins and monsoon shift in the southern, southeastern and northeastern TP."

Line 54: A significant seasonal.

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

"**A significant seasonal** variation of NPF frequency was observed in the TP."

Lines 83-84: The measurements were conducted …

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

"**The measurements were 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)."

Lines 187: the temperature … values … values …

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

"As shown in Fig. 2 and Fig. S9, **the temperature** behavior was characterized by higher **values** in the monsoon season (10.4±4.1 ℃) and lower **values** in the pre-monsoon season (3.1±3.6 ℃) with an average value of 5.3±5.1 ℃."

Lines 191-192: The wind speed …. The wind direction

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

"**The wind speed** (WS) was comparable during the two seasons, which was 4.2±2.7 m s^{-1} in the pre-monsoon season and 4.5±2.7 m s^{-1} in the monsoon season, respectively. **The wind direction** (WD) showed a clear divergence, with westerly and southwesterly winds prevailing in the pre-monsoon season, and southerly winds prevailing in the monsoon season (Fig. S10).

Line 201: … similar to …

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

"On average $PM_{0.8}$ was 1.8±1.0 μg m^{-3}, which was **similar to** PM_1 (2 μg m^{-3}) measured by a high-resolution time-of-flight aerosol mass spectrometer at the Nam Co station in 2015 (Xu et al., 2018a).

Lines 221-222: …lower than that at … comparable with that at …

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

"The frequency at the Nam Co station during the pre-monsoon season was **lower than that at** NCO-P (38%) (Venzac et al., 2008) and **comparable with that** at Mt. Yulong on the southeastern TP (14%) (Shang et al., 2018) in the same season."

Line 276: … photochemical oxidation rate … (be consistent with the text on line 284)

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

"The concentration of sulfuric acid in the atmosphere is related to the degree of SO_2, **photochemical oxidation rate** and CS."

Line 287: … concentrations … levels …

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

"With speculated comparable/lower SO_2 **concentrations** and similar CS and J (O^1D) **levels**, the abundance of gaseous sulfuric acid in NPF days would be approaching, or little lower than that in non-event days."

Line 312: should it rather be: … most probable reasons …

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

"While the concentrations of organic precursors could be the **most probable reasons** for the occurrence of NPF events, the external factors driving the difference in VOC levels between the NPF and non-event days and other conditions that may affect the characteristics of NPF were still unknown."

Lines 316-317: Air pollutants …site are mainly …

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

"Air air pollutants at the observation site **are mainly** brought by air mass transmission."

Line 319: … on non-event days …

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

"It can be found that the dominant air masses **on non-event days** were from the west (almost 100%) and passed by western Nepal, northwest India and Pakistan."

Line 332: … thus triggering …

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

"The summer monsoon can bring the higher organic concentrations in the monsoon season (NPF-monsoon days) compared with those in the pre-monsoon season (NPF-pre and non-event days) (Fig.4), **thus triggering** almost daily NPF events.

Line 378: "in a short time" is a bit vague expression. Please be more specific. I suppose you refer to the few hour or bit more after NPF.

Thanks for the comment. We have reformulated the expressions in the revised manuscript as follows:

"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 **within a few hours after particle nucleation and growth**, that is, the aerosol and CCN production."

II. Reply to Reviewer 2

Reply to Reviewer 2's overall comments:

First of all, I appreciate the tremendous efforts the authors have put into the revised manuscript. Obviously, this manuscript has been significantly improved.

In the interactive discussion, the referees have raised two major concerns: 1) the representativeness of the measurement periods to the pre-monsoon and monsoon seasons; 2) the validation of modeled SO2 and VOC concentrations. Out of them, I think the authors have well addressed the first concern; yet additional evidence is needed for the latter one. I have a few suggestions for the authors' consideration.

We appreciate the comments from the reviewer on this manuscript. We have answered them 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 2's comments (2):

1. The validation of SO2 simulation. There are SO2 data available in Tibet measured at other time, which can be used to validate their model. Note that, the good correlation of SO2 and BC found at another site does not necessarily apply to the location of this study. This is because BC is inert in the atmosphere, while SO2 is quite reactive. Assuming that they are emitted by the same source, the ratio of [SO2]/[BC] would gradually decrease along with photochemical aging, which will deteriorate the correlation.

Thanks for the comment. The comparison between simulated and observed SO_2 at Mt. Yulong on the southern TP has been added to validate the model in the revised manuscript. The statistical metrics of NMB (normalized mean bias) and NME (normalized mean error) values are within the range reported in previous SO_2 modelling result (Mao et al., 2022). The correlation coefficient (R) between simulated and observed SO_2 is 0.44, which reflected that the model can fairly simulate the variation of SO_2 concentration in Tibet.

Mao, J., Li, L., Li, J., Sulaymon, I. D., Xiong, K., Wang, K., Zhu, J., Chen, G., Ye, F., Zhang, N., Qin, Y., Qin, M., and Hu, J.: Evaluation of Long-Term Modeling Fine Particulate Matter and Ozone in China During 2013–2019, Frontiers in Environmental Science, 10, 10.3389/fenvs.2022.872249, 2022.

"**2.3 Model simulation**

For SO_2, the WRF/CMAQ models have been successfully reproduced SO_2 in major regions in China with R of 0.25-0.79 (Mao et al., 2022). And the WRF/CMAQ models achieved good performance in simulating SO_2 at Mt. Yulong on the southern TP (Text S2)."

"Text S2 Model simulation

Model Evaluation

The comparison between simulated and observed SO_2 at Mt. Yulong on the southern TP is shown in Fig. S6,

which helps to validate the model performance. As shown in Table S2, the statistical metrics of NMB (normalized mean bias) and NME (normalized mean error) values are within the range reported in previous SO_2 modelling result (Mao et al., 2022). The correlation coefficient (R) between simulated and observed SO_2 is 0.44, which reflected that the model can fairly simulate the variation of SO_2 concentration in Tibet.

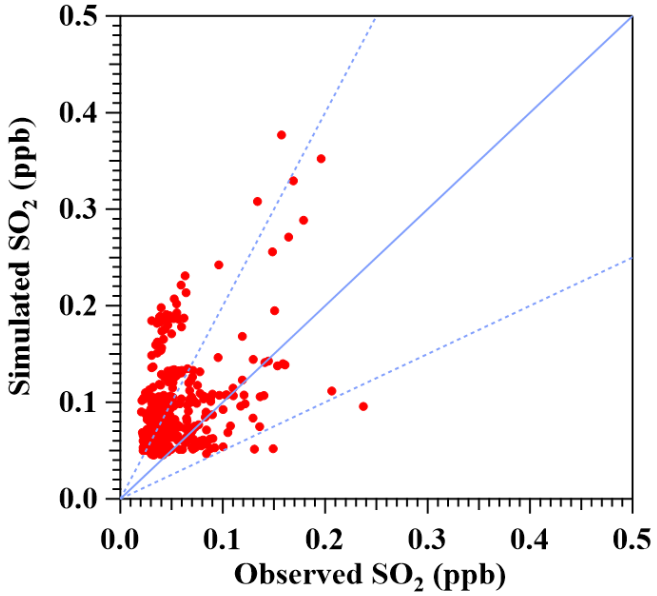

Figure S6. Comparison of simulated and observed SO2 (ppb). SO2 is hourly mean concentration.

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

	PM_1			O_3			VOC			SO_2[a]		
	MFB	**MFE**	**R**	**NMB**	**NME**	**R**	**MFB**	**MFE**	**R**	**NMB**	**NME**	**R**
Statistic	0.49	0.50	0.72	0.14	0.23	0.51	-0.47	0.49	0.41	-0.44	0.50	0.44
Benchmarks	<±0.6	<0.75	>0.4	<±0.15	<0.35	>0.5						
References							<±0.77	<0.74		<±4.38	<±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_3 were suggested by Emery et al. (2017) and Boylan and Russell (2006), respectively. The references for VOC and SO_2 were from Hu et al. (2017) and Mao et al. (2022), respectively.

[a] The statistical metrics for evaluating SO_2 simulation at Mt. Yulong on the southern TP

2. The validation of VOC simulation. I understand that this may be a hard task for the authors. The authors mention that 99 VOCs have been measured during the pre-monsoon season, which covers both NPF days and non-NPF days. The author can further look into these measured VOC, focusing on the comparison of VOC concentrations in NPF and non-NPF days. This would give a good hint. Also, I agree with the referee

that total VOC concentration is not a good quantity, because most VOCs (and especially small VOC molecules) are just spectators of NPF. The authors should pay special attention to VOCs such as monoterpene, sesquiterpene, and heavy aromatics during further analyses.

Thanks for the comment. The comparison of VOC concentrations in NPF and non-NPF days during the pre-monsoon season has been discussed in the manuscript. It is a pity that the monoterpene and sesquiterpene were not measured in this study. The aromatics including benzene, toluene, Styrene and trimethylbenzene were measured in this study. The total concentration of aromatics exhibited a higher level (20%) during the occurrence time of NPF events in NPF-pre days compared with that in non-event days. The aromatics have been considered to contribute substantially to new particle formation (Molteni et al., 2018). The potential NPF precursors such as toluene (Garmash et al., 2020), styrene (Yu et al., 2022) and trimethylbenzene (Molteni et al., 2018) showed higher values in NPF-pre days compared with those in non-event days It gives a good hint for the role of organics in the occurrence of NPF events at the Nam Co station.

Molteni, U., Bianchi, F., Klein, F., El Haddad, I., Frege, C., Rossi, M. J., Dommen, J., and Baltensperger, U.: Formation of highly oxygenated organic molecules from aromatic compounds, Atmos. Chem. Phys., 18, 1909-1921, 10.5194/acp-18-1909-2018, 2018.

Garmash, O., Rissanen, M. P., Pullinen, I., Schmitt, S., Kausiala, O., Tillmann, R., Zhao, D., Percival, C., Bannan, T. J., Priestley, M., Hallquist, Å. M., Kleist, E., Kiendler-Scharr, A., Hallquist, M., Berndt, T., McFiggans, G., Wildt, J., Mentel, T. F., and Ehn, M.: Multi-generation OH oxidation as a source for highly oxygenated organic molecules from aromatics, Atmos. Chem. Phys., 20, 515-537, 10.5194/acp-20-515-2020, 2020.

Yu, S., Jia, L., Xu, Y., and Pan, Y.: Formation of extremely low-volatility organic compounds from styrene ozonolysis: Implication for nucleation, Chemosphere, 305, 135459, https://doi.org/10.1016/j.chemosphere.2022.135459, 2022.

"**3.3.2 Gas precursors**

Due to instrument status, VOC measurement was only available in the pre-monsoon season. The concentration of VOC (total VOC) showed a higher value (20%) during 11:00-18:00 on NPF-pre days compared with non-event days (Fig. 3c). Aromatics, which can be used as the indicator of anthropogenic emissions, also exhibited a higher level (20%) during NPF-pre days (Fig. 3d). The aromatics have been considered to contribute substantially to new particle formation (Molteni et al., 2018). The potential NPF precursors such as toluene (Garmash et al., 2020), styrene (Yu et al., 2022) and trimethylbenzene (Molteni et al., 2018) showed higher values in NPF-pre days compared with those in non-event days (Fig. S15). This suggested that VOC such as aromatics may be the key factor in determining the occurrence of NPF events."

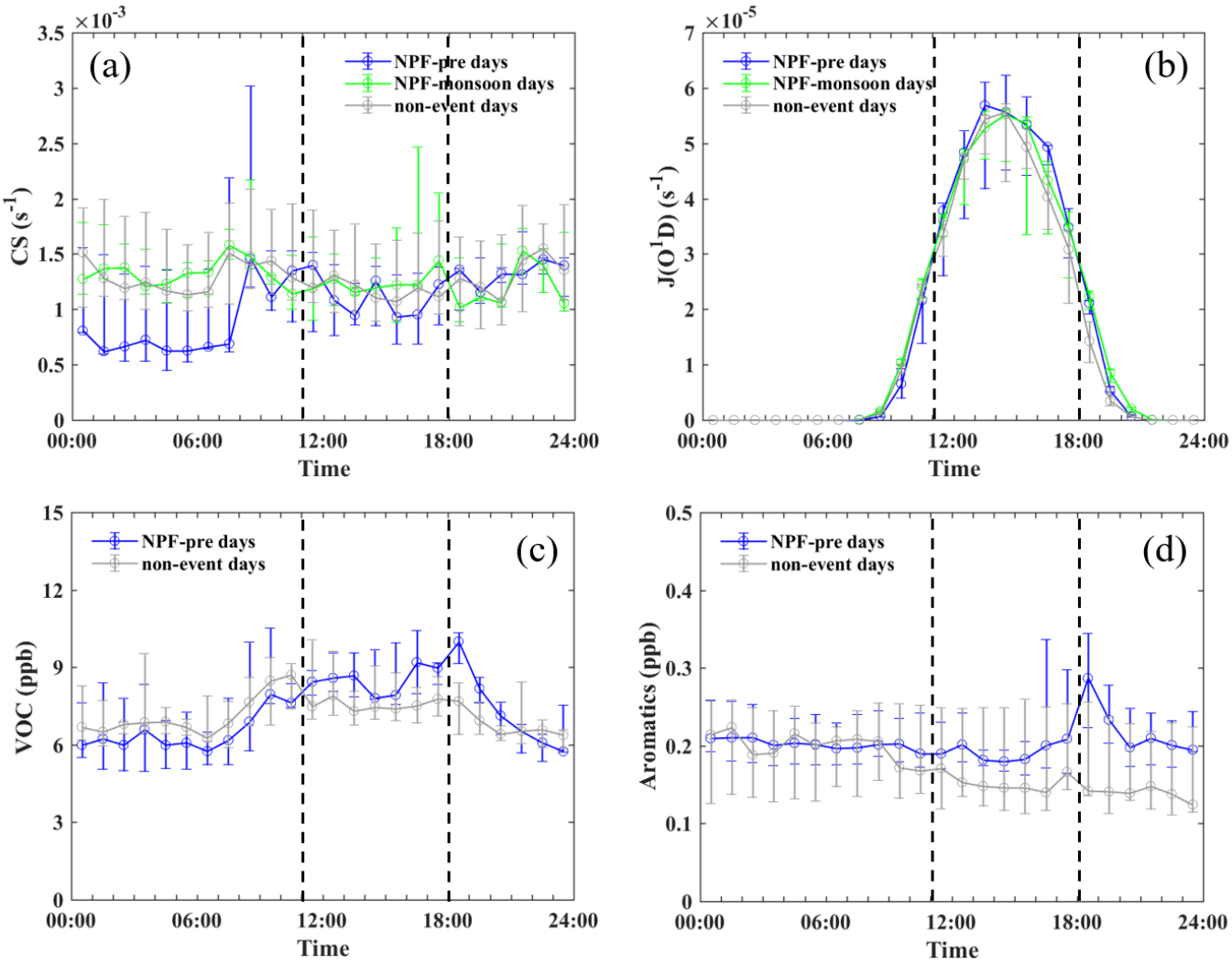

Figure 3. Diurnal variations of (a) condensation sink (CS), (b) JO^1D, the total concentration of (c) VOC and (d) aromatics in NPF-pre days, NPF-monsoon days and non-event days. The upper and lower bars indicate the 75th and 25th percentiles, the markers are the average values.

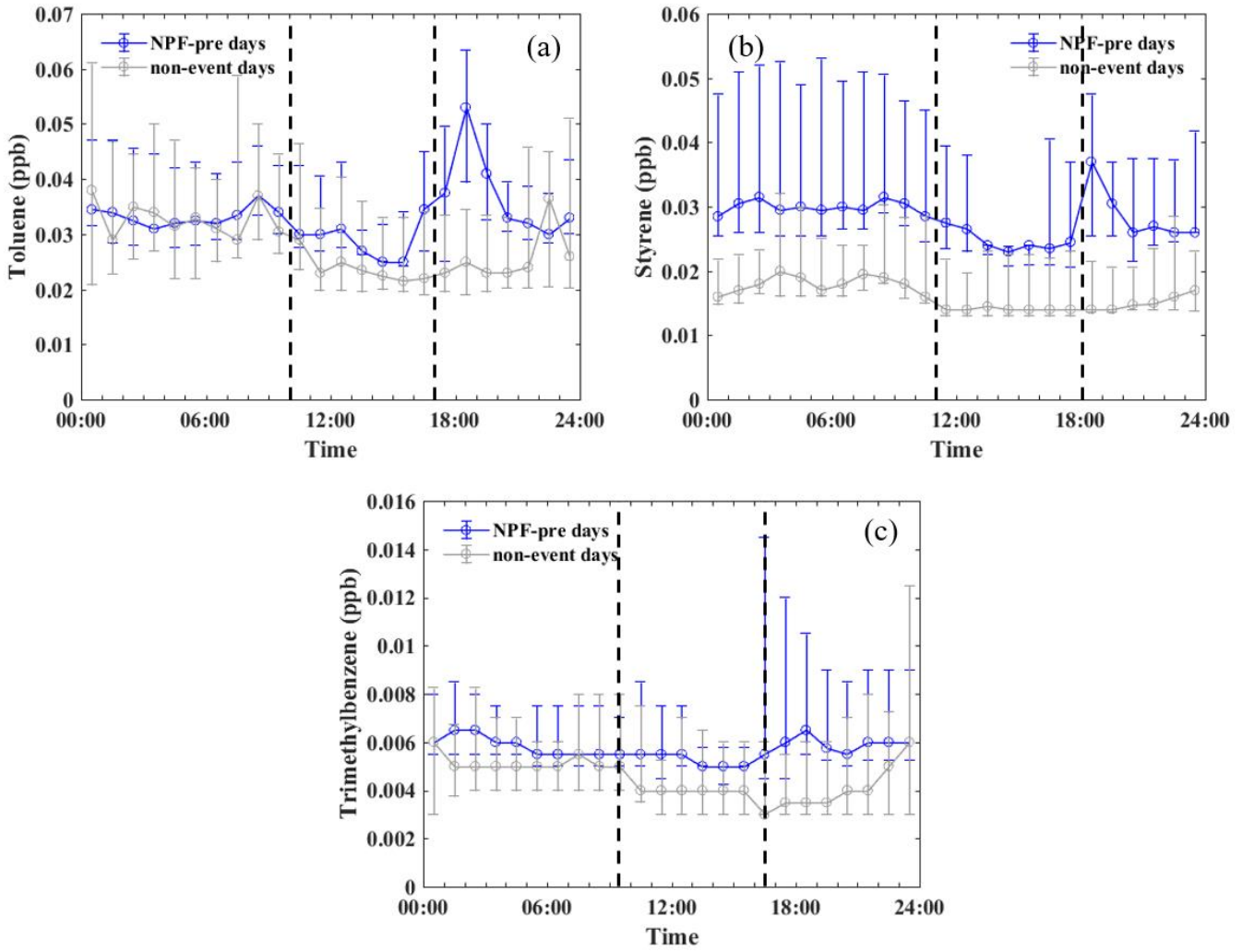

Figure S15. Diurnal variations of concentration of (a) tolunene, (b) styrene and (c) trimethylbenzene in NPF-pre days and non-event days. The upper and lower bars indicate the 75th and 25th percentiles, the markers are the average values."