Dear Editor,

We appreciate the prompt reviews and would like to thank the two reviewers for insightful comments and suggestions on our manuscript entitled "Characteristics of fine particle matters at the top of Shanghai Tower" (MS No.: egusphere-2022-782). We have carefully considered all comments and suggestions. Listed below are our point-by-point responses to all comments and suggestions of this reviewer (Reviewer's points in black, our responses in blue).

Anonymous Referee #2

In this manuscript, the authors gave a very details analysis of observed one year continuous $PM_{2.5}$ and its chemical components at the top of 632 m high Shanghai Tower (SHT). The data collected were precious, and the topic is of great interesting to recognize vertical $PM_{2.5}$ characteristics and its formation processes related to emission, chemical production and boundary layer (BL) etc. The analysis is mostly sound, but some details need clarify. I recommend a minor revision and my specific comments listed below.

Response:

We sincerely thank the reviewer for the valuable comments. These comments have been carefully addressed during revision. Please find our point-to-point response below and highlighted changes in the revised manuscript.

Specific comments:

1. My primary concern is that the study address the $PM_{2.5}$ and its chemical components at SHT dominating by vertical mixing from surface (most in

daytime) and chemical production therein from surface precursors, while omitted the PM originating from transport outside Shanghai. The seasonal winds induced by Asia monsoon are quite difference in upstream (ocean or land, most natural or anthropogenic in background) and could impact much at SHT than on the surface. I suggest the authors should refer to this factor or indicting for future research.

Response:

Thanks for your constructive suggestion. We analyzed the transport pathway at the height of 100 m and 600 m in each season, using 72 h back trajectory from HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Though the two heights had similar tracks (Figure AR1), the small departures might lead to different source origins. Thus, we further calculated the R^2 between $PM_{2.5}$ at SHT and SUR based on hourly and daily-averaged data (Figure AR2). We found that the R^2 increased by 0.12, 0.29, 0.20, and 0.13 on average for spring, summer, autumn, and winter from hourly data to daily-averaged data. The pronounced increase for R^2 indicates that the differences of $PM_{2.5}$ between SUR and 600m mostly came from the subdaily variations.

As suggested, we refer to the influences of regional transport in the conclusion part.

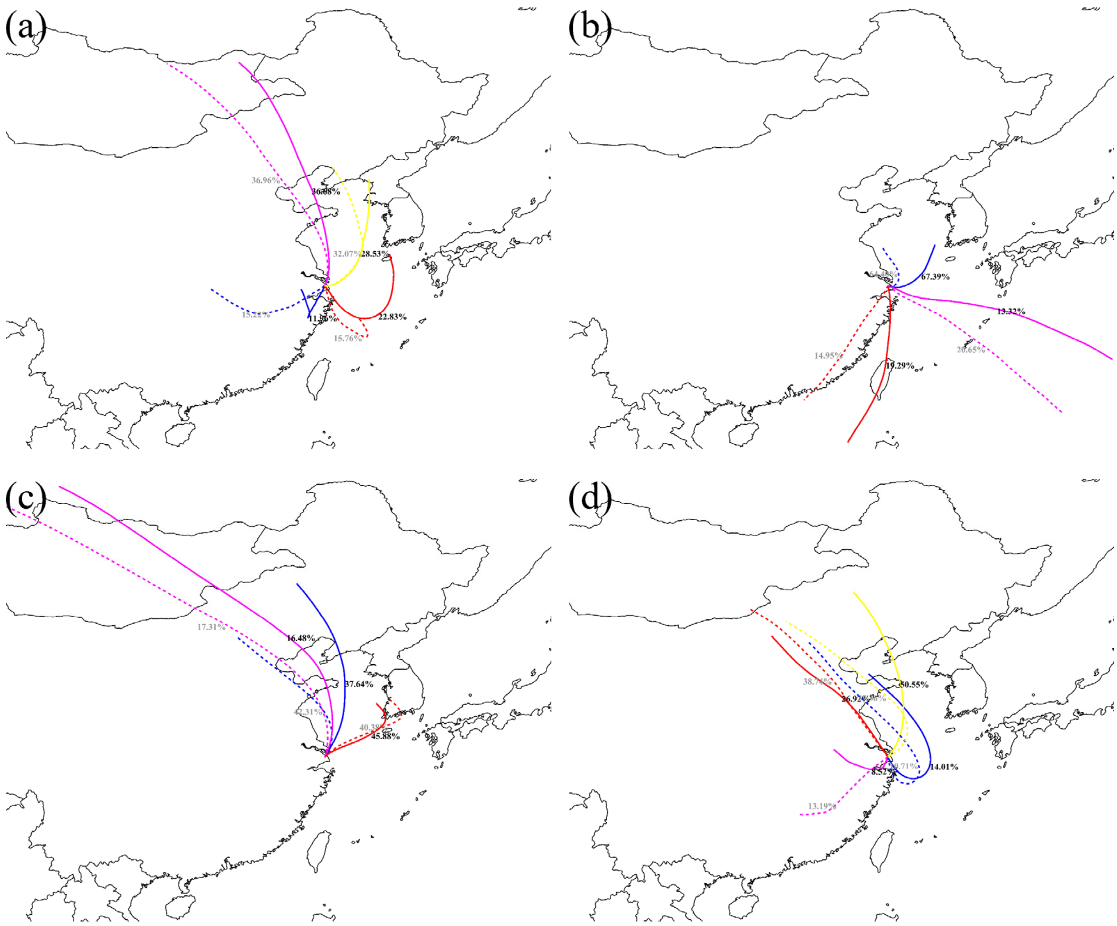

Figure AR1: Air transport pathway at the height of 100 m (solid lines) and 600 m (dashed lines) during spring (a), summer (b), autumn (c), and winter (d). The 72h back trajectory was simulated using HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model with the location of Shanghai Tower as central coordinate.

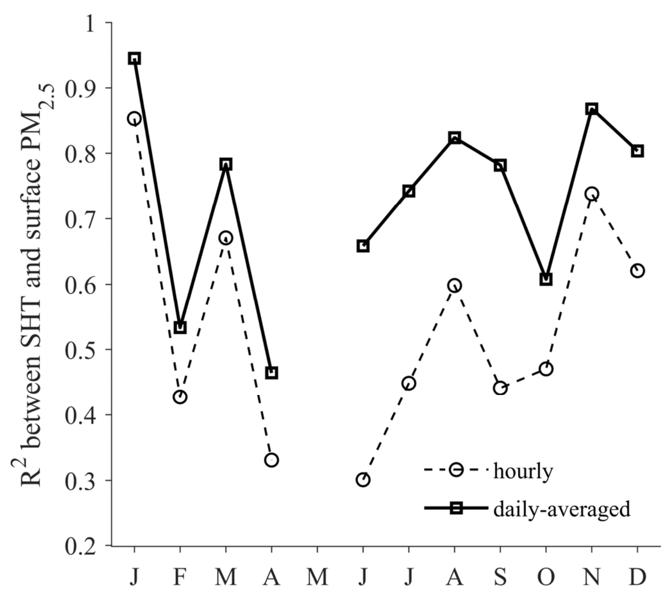

Figure AR2: Monthly variations of correlation coefficients (R^2) between $PM_{2.5}$ at SHT and SUR. The dashed line represents R^2 based on hourly $PM_{2.5}$, and the solid line for daily-averaged $PM_{2.5}$.

2. nitrate (NO_3^-) and sulfate (SO_4^{2-}) should be correctly present in the manuscript.

Response:

Thank you for your suggestion. Though the measurements of nitrate and sulfate are relying on ionized fragments from Q-ACSM, both abbreviates of NO_3 (SO_4) and NO_3^- (SO_4^{2-}) were wildly used in previous studies (e.g., Cao et al., 2019; Zhang et al., 2011; Ng et al., 2011; Zhou et al., 2018). We uniformed the subscripts of nitrate and sulfate as NO_3 and SO_4 throughout the study.

3. In line 69, Shanghai is not only one of the most densely populated megacities in China, but also in the world.

Response:

Thanks for your suggestion. Modifications were made accordingly.

4. In this study, the heights of BL were important. Please give a brief introduction of BLHs in different season and day and night in Shanghai.

Response:

Thanks for your suggestion. We obtained PBL height (PBLH) at SHT from the nearest ERA5 gridded reanalysis data (Hersbach et al., 2020) (https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-singlelevels?tab=form, accessed 27 November 2022). The ERA-PBLH is calculated utilizing a bulk Richardson method, which was widely used for both convective and stable boundary layers (Kim, 2022). According to Wang et al. (2018), the ERA data tend to overestimate PBLH at nighttime, but underestimate PBLH during daytime in Eastern China by comparing with PBLH calculated from radiosonde sounding data. Overall, the reanalysis data can capture the diurnal and seasonal cycle of PBL structure.

As shown in Figure AR3, the autumn found the highest PBLH for its prevailing synoptic of the continental high pressure (characterized as weak winds, strong solar radiation, and dry weather), favorable for the PBL development. PBLH in four seasons presented similar diurnal variations. The PBL started to develop at 06:00-08:00 before reaching a daily top at 13:00-14:00, and then decreased until stabilizing after sunset (18:00-19:00). However, the summertime PBL had the longest development period (06:00-19:00), while the wintertime PBL had the shortest (08:00-18:00). At nighttime, the observatory at SHT generally stood on top of stable BL (SBL) despite the deviations. Whereas the time PBL top reaching SHT site varied during the day. Nevertheless, the PBL had contact with SHT top even for the lower bound of deviation, indicating inevitable mass exchanges between SHT and SUR during the daytime.

Modifications were made accordingly.

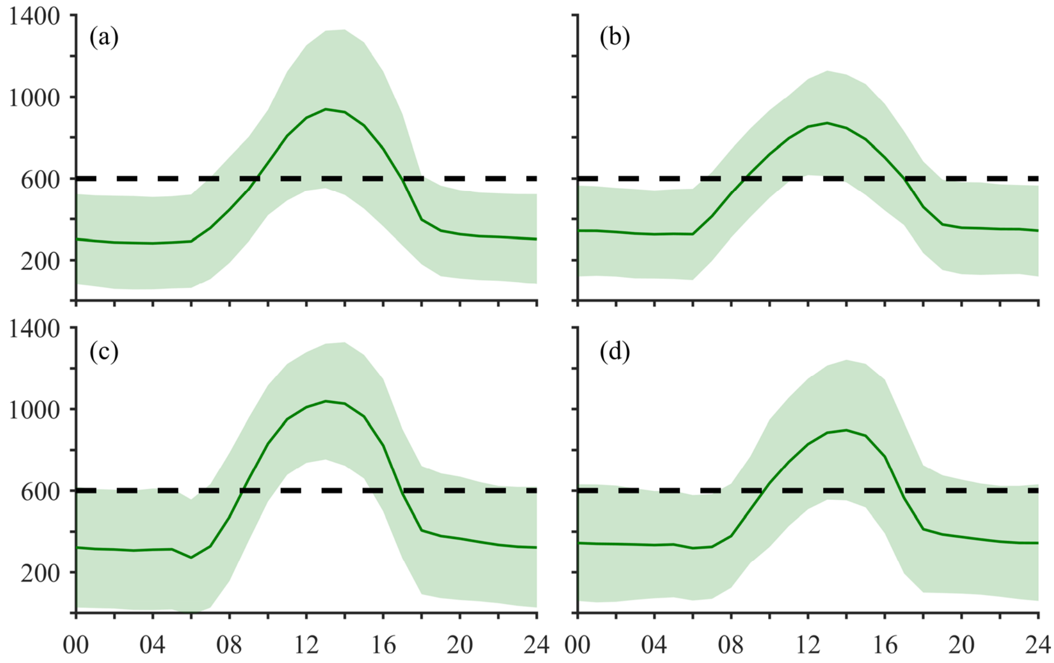

Figure AR3: Diurnal variations of the reanalysis PBL height in spring (a), summer (b), autumn (c), and winter (d) at the grid box where the Shanghai Tower (SHT) site is in. The solid line represents the mean value, and the shaded area stands for the standard deviation. The dash lines represent the altitude (~600 m) of the SHT site.

5. In line 176-178, the inference is not very exact. The seasonal variations of BLH could be key factor for the similar monthly variations of $PM_{2.5}$ at SHT and SUR, and related to regional transport, vertical diffusions etc. And I am happy to find you mentioned of regional transport, while did not raise this in conclusion, abstract and other paragraphs.

Response:

We totally agree that the seasonal variation of BLH was a key factor for the monthly variations of $PM_{2.5}$ at SUR. However, the shallower (deeper) BLH would lead to less (more) mass exchanges between SUR and SHT, resulting in lower (higher) mass concentrations transported from surface to high altitude. Thus, we concluded that the similar monthly variations of $PM_{2.5}$ at

SHT and SUR were more likely related to regional transport and local emissions.

6. In 188-190, the anomalies may reflect the seasonal variations of BLHs.

Response:

Thanks again for your suggestions regarding seasonal behaviors of PM. As mentioned in question 5, the seasonal variations of BLHs had opposite impacts on SHT and SUR. However, the shallower BLH meant less contact time between SHT and SUR air in winter, presenting larger differences between the anomalies at two altitudes.

7. In 210-211, "completely" is not very exact because in some synoptic conditions, the mass and energy exchange between free troposphere and within the BL could occur.

Response:

Thanks for noting. We changed "completely" to "mostly".

8. What were the definitions of POA, OOA, and HOA, and their chemical components in this study?

Response:

The POA and OOA were two factors we retrieved from PMF analysis. As the profile of POA was a mixture of HOA, COA, and CCOA features, which were identified as primary components (Duan et al., 2019). the HOA profile is recognized by noticeable hydrocarbon ion series of C_nH_{2n-1} and C_nH_{2n+1}; particularly m/z 27, 29, 41, 43, 55, 57, 67 and 71. COA is characterized by higher ratio of m/z 55 than m/z 57, and CCOA mass spectrum is acknowledged as distinctive polycyclic aromatic hydrocarbons (PAHs) fragments. OOA profile sees prominent ion fragment at m/z 44 (CO_2^+). Modifications were made accordingly.

9. In figure 5, why there was the largest difference of $PM_{2.5}$ between SHT&SUR in summer, while the largest difference of NOR in winter and spring in figure 7?

Response:

Thanks for your suggestion. We gathered Monitor for AeRosols and Gases in ambient (MARGA) data from Pudong New District Environmental Monitoring Station to calculate the NOR at surface. As the largest differences of $PM_{2.5}$ between SHT and SUR were found around noon, the NOR during the daytime were calculated accordingly. As shown in Table AR1, the NOR was higher at SHT than SUR in spring, summer, and autumn, while lower at SHT in winter. Besides, the most significant difference of NOR appeared in summer, when the largest $PM_{2.5}$ departures between two altitudes were found.

Table AR1: The NOR during the daytime at SHT and SUR in four seasons. The NOR was calculated as: $[NO_3]/([NO_3]+[NO_2])$, where [x] points to the molar concentration of x.

	Spring	Summer	Autumn	Winter
SHT	0.13	0.10	0.09	0.15
SUR	0.12	0.07	0.07	0.16

10. In line 370, latitude should be altitude.

Response:

Thank you for the note. Revised.

11. In line 374, "since the SO_2 level was relatively lower than the other seasons.". or also because the favorable diffusion and wet scavenging condition of atmosphere in summer.

Response:

Thank you. It was revised as suggested.

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