Response to Anonymous Referee #3's Comments

General comments

This study reports a dust event with high relative humidity and low wind speeds occurred in Shanghai during the period of October 29-November 2, 2019. The dust event was divided into three obvious stages, of which the first stage was a dust invasion stage, the second stage was a dust development period, and the third stage was a dust backflow period. Meanwhile, chemical characteristics of aerosols in the three stages were investigated, and a simplified method was deployed to identify and estimate the amounts of major aerosol species from transport and secondary formation. The study method is reasonable, data is reliable, and conclusion is credible. But there existed a lot of aspects to be revised and improved in the manuscript. Written language and logical relationship need to be improved. I suggest to consider the paper for publication after a major revision.

We sincerely thank for the reviewer's positive comments and helpful suggestions on this manuscript. Based on the specific comments, we have responded to all the comments point-by-point and made corresponding changes in the manuscript as highlighted in red color. We feel the revisions based on the reviewer's comments have greatly improved the quality of this manuscript. Please check the detailed responses to all the comments as below.

Specific comments

1. In abstract, L28, the phrase "by with high concentrations of particulate matters but relatively short duration" should be changed to "by high concentrations of particulate matters but relatively short duration".

Thanks for pointing out this typo. The word "with" is now deleted in the revision.

2. In introduction, L121, the sentence "In contrast, this study aims to depict an atypical

dust event was observed in Shanghai, a coastal mega-city in Eastern China." is error.

This is indeed a grammatical error. The sentence is now revised as "In contrast, this study aims to depict an atypical dust event that was observed in Shanghai, a coastal mega-city in Eastern China.".

3. In section 2.2, all the online instruments used in this study should be normally calibrated so as to guarantee data quality, therefore, normal calibrations of these monitoring instruments should be added in the section.

Thanks for the suggestion. For all the online instruments equipped at the Pudong supersite, they are routinely maintained by professional technicians. The online data are checked each day via an online integrated information system. Thus, the data quality can be assured. In Line 182 of the revised manuscript, the sentence "All instruments are routinely maintained and calibrated to ensure the quality of data." is added.

4. In sections 2.3 and 2.4, the models used in this study should be detailed.

In Line 187-205, Sections 2.3 is revised as below.

The ISORROPIA II model is subject to the principle of minimizing the Gibbs energy of the multi-phase aerosol system, leading to a computationally intensive optimization problem (Song et al., 2018). The model can predict the physical state and compositions of atmospheric inorganic species (NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}, SO_4^{2-}, NO_3^- and Cl^-) with their gas- and particle-phase concentrations and meteorological parameters (relative humidity and temperature) as model inputs. The model includes two modes, i.e., reverse and forward mode. The reverse mode calculates the equilibrium partitioning based on aerosol-phase concentrations only, while the latter uses both aerosol-phase and gas-phase concentrations as inputs. Moreover, particles can be assumed as "metastable" with liquid-phase but no solid participating while "stable" with the liquid and solid phases or both. The ISORROPIA running in the forward mode at the metastable state was applied in this study. Aerosol pH was calculated based on the equilibrium particle hydronium ion concentration and aerosol liquid water content (ALWC) obtained from model results. The performances and advantages of

ISORROPIA over the usage of other thermodynamic equilibrium codes has been assessed in numerous studies (Nenes et al., 1998; West et al., 1999; Ansari and Pandis, 1999; Yu et al., 2005).

In Line 209 – 221, Sections 2.4 is revised as below.

The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) was used to compute the backward trajectories of the air parcels during the dust events. It is a widely used model that computes dispersion following the particle or puff. The advection of a particle or puff is computed from the average of the three-dimensional velocity vectors for the initial position and the first-guess position (Draxler and Hess, 1998). Turbulent velocity components are expressed as a function of the velocity variance, a statistical quantity derived from the meteorological data, and the Lagrangian time scale. The calculation of air mass trajectories can be used to depict the airflow patterns for interpreting the transport of air pollutants over various spatial and temporal ranges (Stein et al., 2015). In this study, the HYSPLIT model was driven by meteorological data outputs from the Global Data Assimilation System (GDAS) (Su et al., 2015), which is available at ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1. Air mass trajectories were launched at different heights from the ground and a total duration of 48 hours simulation was conducted.

References:

Draxler, R. R., and G. D. Hess, 1998: An overview of the HYSPLIT_4 modeling system for trajectories, dispersion, and deposition. Aust. Meteor. Mag., 47, 295–308.

Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., Ngan, F., NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System, Bulletin of the American Meteorological Society, 2015, 2059–2077.

5. In section 2.5, the sentence “Initially, the quasi-first-order reaction rate constant for heterogeneous conversion from NH3 to NH4+ (khet, s-1) is calculated according to (Liu et al., 2022).” is incomplete. In addition, the two formulas should be numbered.

All the formulas in this study should be numbered in turn.

This sentence is revised as "Initially, the quasi-first-order reaction rate constant for heterogeneous conversion from NH_3 to NH_4^+ (k_{het}, s^{-1}) is calculated by Eq. (1) (Liu et al., 2022)." in Line 228.

All the formulas in this study have been numbered in the revision.

6. In section 2 methodology, sampling duration should be supplemented.

In the revision, we have added the sampling duration for the applied instruments as below.

Line 162-163: Samples were collected for 40 min and then analyzed in the following 20 min.

Line 171-172: These parameters were measured at the temporal resolution of 5min.

Line 174: Meteorological parameters (ambient temperature, relative humidity, wind speed, and wind direction) were obtained by a Vaisala Weather transmitter (WXT520) at the temporal resolution of 1min.

Line 175-181: The height of planetary boundary layer (PBL) was retrieved from a ceilometer (CL31, Vaisala) at the temporal resolution of 30 min. Vertical profiles of aerosol optical properties were obtained by an aerosol lidar (AGJ, AIOFM) at the temporal resolution of 30 min and vertical resolution of 7.5 m, respectively. Vertical profiles of ozone were obtained by an ozone lidar (LIDAR-G-2000, WUXIZHONGKE) at the temporal resolution of 15 min and vertical resolution of 7.5 m, respectively.

7. In section 3.1, L216-218, the sentence "From October 25 to 28, the mean wind speed remained relatively low of 0.9±0.72m/s with a peak value of 3.1m/s, and predominantly blowing from the northwest." is suggested to be changed to "From October 25 to 28, the mean wind speed was 0.9±0.72m/s with a peak value of 3.1m/s, remaining relatively low, and predominantly blowing from the northwest."

Thanks for the suggestion. It is revised in Line 244-245 as suggested.

8. L218, the sentence "The mean concentration of PM2.5 and PM10 was 34.7 and 44.2

μg/m3, respectively.” should be revised to “The mean concentrations of PM2.5 and PM10 were 34.7 and 44.2 μg/m3, respectively.”

It is corrected in Line 247 as suggested.

9. L214-228, in this paragraph dust and non-dust periods should be identified, but authors did not give related discussion.

In this paragraph, the definitions of dust and non-dust periods have been added in Line 255-263.

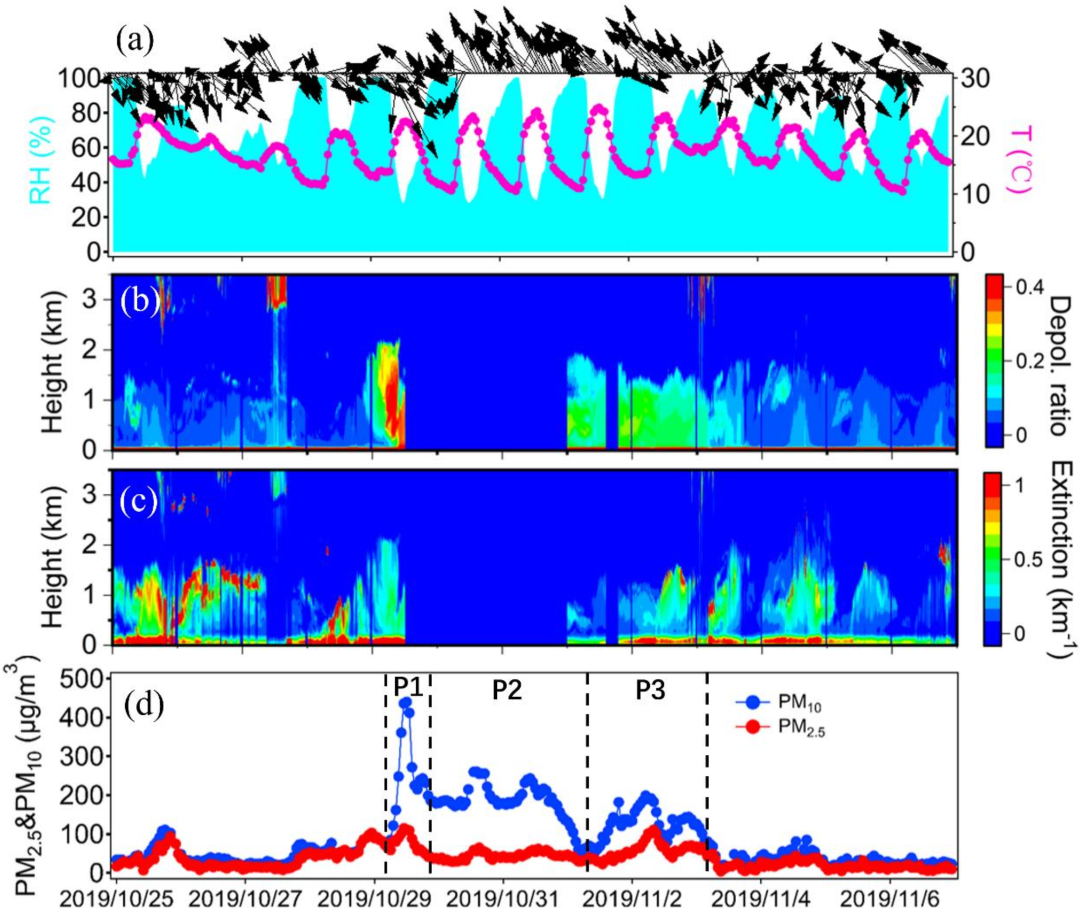
By using the $PM_{2.5}/PM_{10}$ mass ratio of 0.4 as a threshold (Fan et al., 2021), the period from October 29 to November 2 was defined as the dust period in this study. The remaining days, including October 25 to October 28 and November 3 to November 6, were defined as the non-dust period. Throughout the entire dust period, the mean concentrations of $PM_{2.5}$ and PM_{10} reached $53.3 \pm 20.5 \mu g/m^3$ and $172.4 \pm 70.2 \mu g/m^3$, respectively, yielding a low $PM_{2.5}/PM_{10}$ ratio of 0.34 ± 0.15. As a comparison, $PM_{2.5}$ and PM_{10} during the non-dust period was $38.9 \mu g/m^3$ and $49.8 \mu g/m^3$, respectively, exhibiting a relatively high $PM_{2.5}/PM_{10}$ ratio of 0.62 ± 0.20.

Reference:

Fan, H., Zhao, C., Yang, Y., and Yang, X.: Spatio-Temporal Variations of the PM2.5/PM10 Ratios and Its Application to Air Pollution Type Classification in China, Frontiers in Environmental Science, 2021, https://doi.org/10.3389/fenvs.2021.692440.

10. In Figure 1, P1, P2 and P3 should be put in Figure 1d and separated with vertical lines.

As suggested, Figure 1 is revised as below.



11. What did aerosol depolarization ratio was used to explain? Please explain correlation between the depolarization ratio values and the dust event and impacts of relative humidity on the depolarization ratio.

The depolarization ratio, a measure of the irregularity of the scatterer shape, is the most important property of dust measured by lidar systems (Shimizu et al., 2017). The high depolarization ratio of aerosol was due to the nonsphericity (irregular shapes) and relatively large size of particles (Mcneil and Carswell, 1975). If the depolarization ratio of the region is less than 10% it is identified as spherical aerosol, and if it exceeds 10% it is identified as mineral dust (Shimizu et al., 2004).

In general, if relative humidity is high, particles can absorb more water and thus become more spheric, thus lowering the depolarization ratio, and vice versa.

In the revision (Line 251 - 254), we have made changes to describe more clearly about the depolarization ratio.

In general, if the particle depolarization ratio exceeds 10%, the aerosol is identified as mineral dust (Shimizu et al., 2004) due to the nonsphericity (irregular shapes) and relatively large size of particles (Mcneil and Carswell, 1975).

References:

Shimizu, A., Nishizawa, T., Jin, Y., Kim, S. W., Wang, Z., Batdorj, D., and Sugimoto, N.: Evolution of a lidar network for tropo-spheric aerosol detection in East Asia, Opt. Eng., 56, 031219, https://doi.org/10.1117/1.oe.56.3.031219, 2017.

Shimizu, A., N. Sugimoto, I. Matsui, K. Arao, I. Uno, T. Murayama, N. Kagawa, K. Aoki, A. Uchiyama, and A. Yamazaki (2004), Continuous observations of Asian dust and other aerosols by polarization lidars in China and Japan during ACE-Asia, J. Geophys. Res.,109, D19S17, doi:10.1029/ 2002JD003253.

Mcneil, W. R. and Carswell, A. I.: Lidar Polarization Studies of Troposphere, Appl. Opt., 14, 2158–2168, 1975.

12. L237-244, “In this study………non-dust period”, please supply specific start and end time.

In Line 257-259, the non-dust period is defined as “October 25 to October 28 and November 3 to November 6”.

13. Huang et al., 2010a and Huang et al., 2010b were the same reference. Please check the reference.

Thanks for pointing out this mistake. It is corrected in the text and reference section.

14. L337-342, in “Firstly, …... ~1 ppbv/h (Wang et al., 2020).”, related explanation is lack of logic. Please think it over and revise the explanation. In the text, there are many similar logical problems need to be further checked and revised.

This paragraph is revised in Line 375-380 as below.

Firstly, the mean wind speed was low of 0.4 and 0.6 m/s during P2 and P3, respectively.

One numerical study conducted during the similar period suggested that the low wind speed caused reduction of boundary layer height and the warming of the lower atmosphere, thus accelerating the ozone formation by ~1 ppbv/h (Wang et al., 2020). Consequently, this weak synoptic system was favorable for the accumulation of ozone.

15. L345, Figure 5b here should be Figure 4b.

Thanks for pointing out this typo. It is corrected in the revision.

16. In correlation heatmaps, please explain the meaning of dot size.

The size of dot means the value of the correlation coefficient, corresponding to the numbers in the heatmaps. Also, the black star inside the dot means the correlation is significant ($p<0.05$). In the caption of Figure 7, the meaning of dots is added in Line 503-507.

17. L494, how to understand the sentence "both SO42- and NO3- showed moderate to significant correlations with Na+."

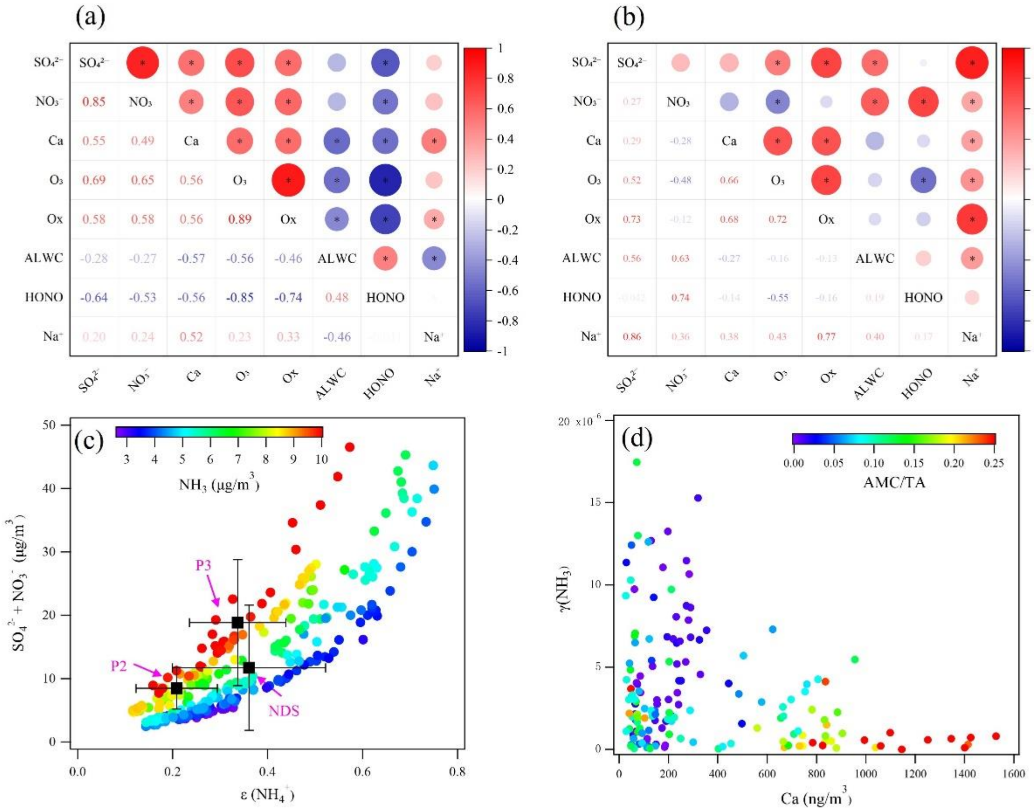
This sentence is based on the correlation analysis as shown in Figure 7b. We meant to say that by using Na^+ as the tracer of sea salts, the moderate to significant correlations between SO_4^{2-}/NO_3^- and Na^+ indicated the contribution of sea salts to the secondary aerosol formation during the dust backflow over the ocean.

In the revision (Line 539), we have stated more clearly as below.

In addition, unlike P2, both SO_4^{2-} and NO_3^- showed moderate to significant correlations with Na^+, a tracer of sea salts (Figure 7b).

18. The mean states of P2, P3 and NDS illustrated in Figure 7c were unclear, hope to better present these states.

In the revised Figure 7c, the mean states of P2, P3 and NDS have been illustrated more clearly as below. In addition, the meaning of P2, P3 and NDS is added in the caption of Figure 7.



19. In section 3.5, in the formula TPPD,i = AVLYG,i × (1-k), please explain the meaning of “1-k ”.

k *is* defined as the removal coefficient of aerosols during the dust transport. Thus, 1-k means the transport fraction of aerosols. In the revision, the meaning of “1-k” is explained in Line 623-624.

20. The conclusions need to be further condensed.

Thanks for the suggestion. Some redundant writings are removed. Please check Line 651-705 for the changes.

Response to Anonymous Referee #4's Comments

The quality of the revised manuscript is significantly improved compared to the original version. The authors adequately addressed the questions raised by the reviewers. Overall, I support the publication of the manuscript upon addressing the following questions.

We sincerely thank again for the reviewer's further assessment on this manuscript. Based on the specific comments, we have responded to all the comments point-by-point and made corresponding changes in the manuscript as highlighted in red color. We feel the revisions based on the reviewer's comments have greatly improved the quality of this manuscript. Please check the detailed responses to all the comments as below.

(1) For the analysis of sulfate formation, I'm still not convinced. In several places (i.e., Lines 473-475 and Lines 623-625), the authors suggested sulfate showed correlation with O3 or O3+NO2, but not with ALWC. They also suggested gas-phase oxidation dominate the sulfate and nitrate formation in Lines 623-625. I'm not sure which mechanism the authors were trying to refer to. SO4 can only be efficiently formed through aqueous oxidation of SO2 by O3, which would need aerosol water. Please clarify.

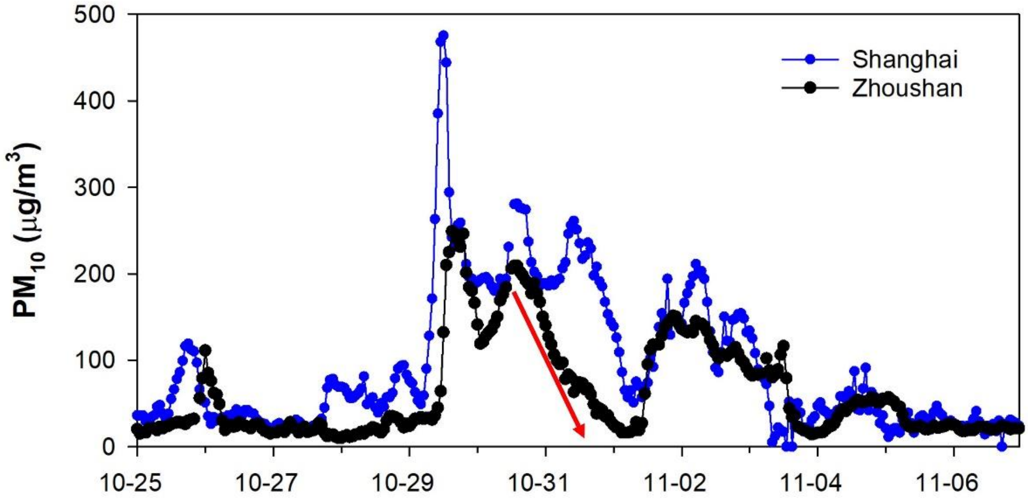
Thanks for the comments and we quite agree that the reaction of SO_2 with O_3 is quite slow without the participation of aerosol water. Although the observational data shows correlation between sulfate with and or O_3+NO_2, but not with ALWC, it doesn't preclude the role of aerosol liquid water in the formation of sulfate. It was simulated by the ISORROPIA model that ALWC reached a moderate level of 24 μg/m^3 during P2. Thus, aerosol liquid water should still be an important medium for the aerosol formation. However, based on the data analysis, we think the sulfate pathways in the aqueous phase such as oxidation by H_2O_2, catalysis by tracer metals, and oxidation by NO_2 should be overwhelmed by the sulfate pathway of SO_2 oxidized by O_3.

To make this statement more clearly, the related sentence is revised in Line 518-521 as below.

During P2, both SO_4^{2-} and NO_3^- displayed the most significant correlations with O_3 and Ox (O_3+NO_2), while even negatively correlated with ALWC. In regard of the obvious ozone enhancement phenomenon as discussed in Section 3.3.1, the photochemistry pathway for the secondary aerosol formation (e.g., S(IV) + O_3 (aq) → S(VI)) should overwhelm over the aqueous phase pathways, e.g., oxidation by H_2O_2, catalysis by tracer metals, and oxidation by NO_2.

(2) Lines 547- 548, I don't quite understand the calculation in section 3.5. The authors suggested the dust plume in P1 were blown into the oceanic area followed by a mixing period (P2, Lines 260-271). The dust plume in P1 might linger over coastal area. This means the backflow may involve both the dust originally blown out in P1 as well as the dust from Lianyungang. The current calculation of "k" only considers the dust transported from Lianyungang. And the so-called background BC concentration in Pudong cannot represent the BC concentration in the dust plume that lingering over the ocean.

Thanks for the comments. It is a very good point that there is indeed possibility of lingering dust over the ocean that could be also mixed with the dust from Lianyungang during the backflow. The reliability of the developed method depends on the abundances of lingering dust over the ocean. However, there are no observational data over the ocean for the assessment. In this regard, we used observational data in Zhoushan archipelago for additional analysis in the figure below.



The time-series of PM_{10} observed at both Shanghai and Zhoushan are compared. It can be seen that PM_{10} quickly decreased to very low concentrations after the strong dust during P1. This is a common phenomenon that strong dust plumes quickly dissipated due to the strong cold fronts. It should be noted that Zhoushan archipelago is located less than 100km from the coasts. Based on the transport trajectories shown in Figure 2f, the P3 dust plumes were much far away from the coasts. Thus, it could be reasonably conjectured that the lingering dust over the ocean was limited.

However, we do agree with the reviewer that there could be interference from the lingering dust over the ocean. In the revision, we have added a paragraph in Line 639-643 as below.

It should be noted that the simple method devised in this study may have inherent uncertainties. Considering the prolonged duration of the dust event, it is possible that certain dust particles lingered over the open ocean. Consequently, the contributions attributed to aerosol transport should be considered as a conservative estimate or lower bound, rather than an exhaustive assessment.