Reponses to referee(s) comments

Dear Editor,

Thank you for your efforts for handling our manuscript. We appreciate to receive the useful comments from all referees. These comments are very constructive, and we have now further revised our manuscript in light of all referees' comments. Based on the helpful suggestions from all reviewers, we believe that we should have addressed questions and concerns from all referees appropriately, and adequately. Please find our point-by-point responses below.

Anonymous Referee #1:

The manuscript has been improved, but still need further revision to meet the standard of ACP. Some of my concerns were not well addressed.

Response: Thank you for your valuable time to review this manuscript. We have further revised the manuscript based on your constructive comments.

1. "The number of synoptic patterns (k) is optimized when the ΔECV is at the highest value, which suggests that the performance of classification has been improved substantially and with stability." The authors presented the explained cluster variances from 4 types to 15 types. How about 2 or 3 types? The results can have a higher increment of the ECV? The highest value of ΔECV is no guarantee of reliable classifications. More in-depth analysis and discussions on the 4-type classification results may be added, as well as its uncertainties and limitations. A specific synoptic pattern can be caused by the seasonal movement of WPSH or the quick pass of a typhoon, which can lead to different atmospheric processes (e.g. precipitation, LLJ, large-scale subsidence) and pollution levels.

RESPONSE: Many thanks for your valuable suggestions. An important criterion to determine the number of SWPs is to ensure that the differences between different synoptic patterns are the largest, while the differences within the same synoptic pattern is the smallest. ECV is usually recommended as an indication, as a greater ECV value often corresponds to a better performance of the synoptic pattern classification (Hoffmann and Heinke SchlüNzen, 2013). The highest value of ΔECV means that the performance in the synoptic pattern classification is improved substantially (Ning et al., 2019). Therefore, both higher ECV and ΔECV values were considered in our study. We found the small value of ECV when the number of SWPs was two or three, indicating greater differences within the same synoptic pattern. The ECV value showed the highest increase when the number of SWPs was four, which means the differences within the same synoptic pattern was significantly improved (Fig. S1). Therefore, four SWPs were finally selected in our study. We have added the above analysis in the text S1 of supplementary material. We have added more discussions about classification results, uncertainties and limitations at lines 471–480 on page 17. Please also see as follow:

"It is important to note that our work contains a few limitations and uncertainties. Although T-PCA, an objective classification method, was chosen in this study, there were still some subjective decisions made, e.g., the number of SWPs (Huth et al., 2008). In the present work, we selected four SWPs based on both the larger ECV and greater ΔECV to furthest reduce the subjective impact. Nevertheless, at a large scale, the present four SWPs were closely associated with intraseasonal movements of the WPSH, because the WPSH is one of the most

important components of the present large-scale SWPs in summertime (Zhao and Wang, 2017). In addition, note that short-term disturbances induced by typhoons with specific pattern were not excluded. The quick passage of a typhoon in summer could lead to various atmospheric processes (e.g., precipitation, large-scale subsidence) and pollution levels (Deng et al., 2019), which should be explored in future work."

2. The detailed descriptions of typhoon-case (Fig. R1 and R2) can be added in the revised manuscript to help readers to understand the sharp movement of WPSH.

RESPONSE: We appreciate your kind suggestion. We have added these at lines 295–300 on page 11. Please also see as follow:

"For instance, tropical storm NEPARTAK generated at 0000 UTC (0800 BJT) 3 July 2016 over the western North Pacific and upgraded to a super typhoon at 1200 UTC (2000 BJT) 5 July 2016 (Fig. S5; see also Su et al., 2017). Due to the rapid movement of NEPARTAK to the northwest, the WPSH quickly decomposed a monomer and moved north. With the strengthening and landing of the typhoon, the monomer gradually collapsed. The SWP also underwent a transition from Type 2 to Type 4, and then to Type 1 (Figs. 4 and S5).".

Reference:

Su, H., Qian, C., Gu, H. and Wang, Q.: The Impact of Tropical Cyclones on China in 2016, Trop. Cyclone Res. Rev., 5(1–2), 1–11, doi:10.6057/2017TCRRh1.01, 2017

3. The ERA5 data were used in this study, but not described in the manuscript. Why not classify the 500-hPa fields of ERA-5, and then carefully analyzed the PBL and precipitation based on the hourly ERA-5 data. How about the consistencies/differences between the ERA-5 data and NCEP data.

Response: Sorry for our negligence. The description of ERA5 data have been added at lines 175–180 on page 7 as follow: "For further analysis of the modulation of the co-occurrence of O_3–$PM_{2.5}$ pollution by the boundary layer structure in some local areas, we also used the BLH, uv-wind, vertical velocity, RH and temperature fields of the fifth generation European Centre for Medium-Range Weather Forecasts reanalysis (ERA5), which has a high spatiotemporal resolution (0.25° × 0.25°, hourly; https://cds.climate.copernicus.eu/cdsapp#!/home)."

Our original intention was to explore the regulation of large-scale synoptic circulation on compound pollution. Guan and Li (2021) proposed that the correlation coefficient of geopotential height between NCEP reanalysis data and scientific experimental data (the third Qinghai–Tibet Plateau atmospheric science experiment from 2015 to 2017) is above 0.99. Consequently, we selected geopotential heights from NCEP data as a categorical variable. For further analysis of the modulation of the co-occurrence of O_3–$PM_{2.5}$ pollution by the boundary layer structure, we used ERA5 data (such as hourly BLH, temperature data, etc.) with a high spatiotemporal resolution as well. In order to strengthen the robustness of our work, we provided a figure of four SWPs based on ERA5 reanalysis data (Fig. S2), which is highly consistent with NCEP reanalysis data at large scales (Figs. 4 and S2). Additionally, we also furtherly compared the differences between NCEP and ERA5 data. As shown in Fig. S3, the geopotential height of NCEP reanalysis data is significantly positively correlated with that of ERA5 data. Especially in eastern China, the correlation

coefficient between the two is greater than 0.96, and all of our classification areas have passed the 99% level of significance test. Overall, the results of this study are robust. We have inquired into the influence of local boundary layer structure on compound pollution events based on the hourly PBL and other meteorological variables of ERA5 data. This has deepened our understanding of the mechanism of the compound pollution events in eastern China during summertime.

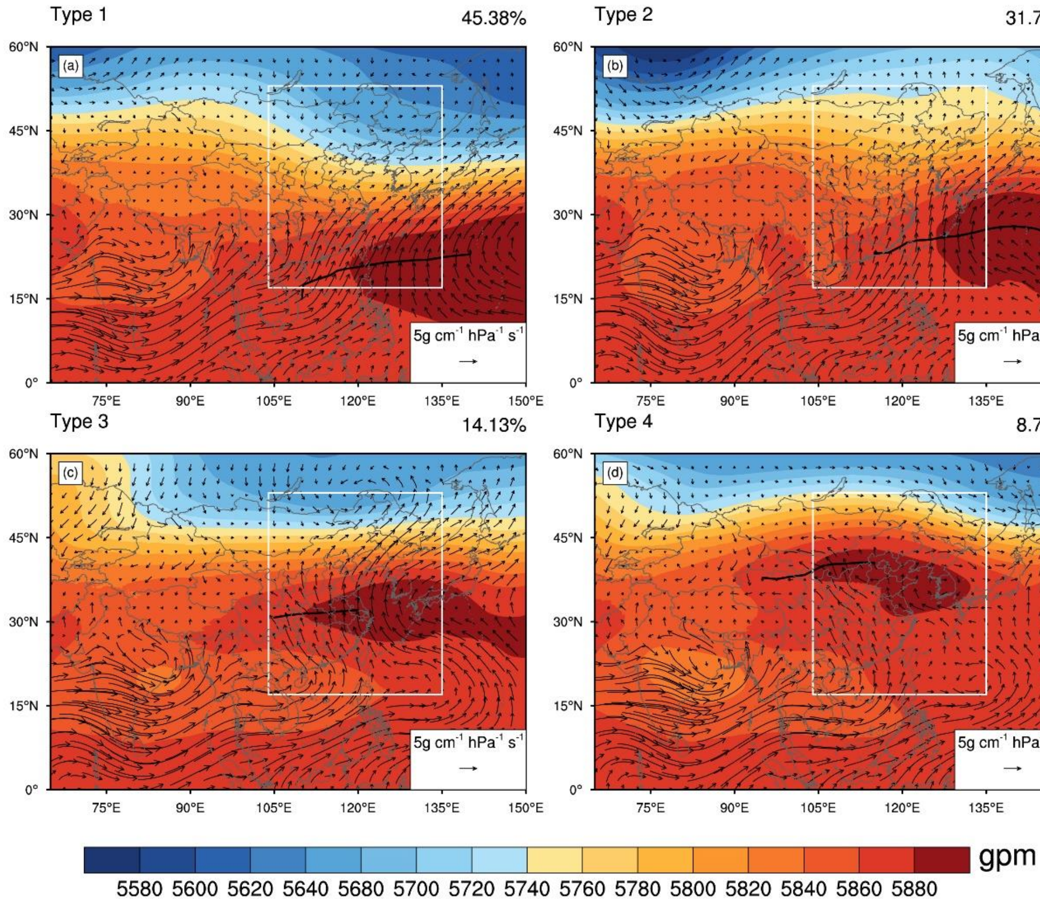

Fig. S2. As in Fig. 4 but for ERA5 reanalysis data.

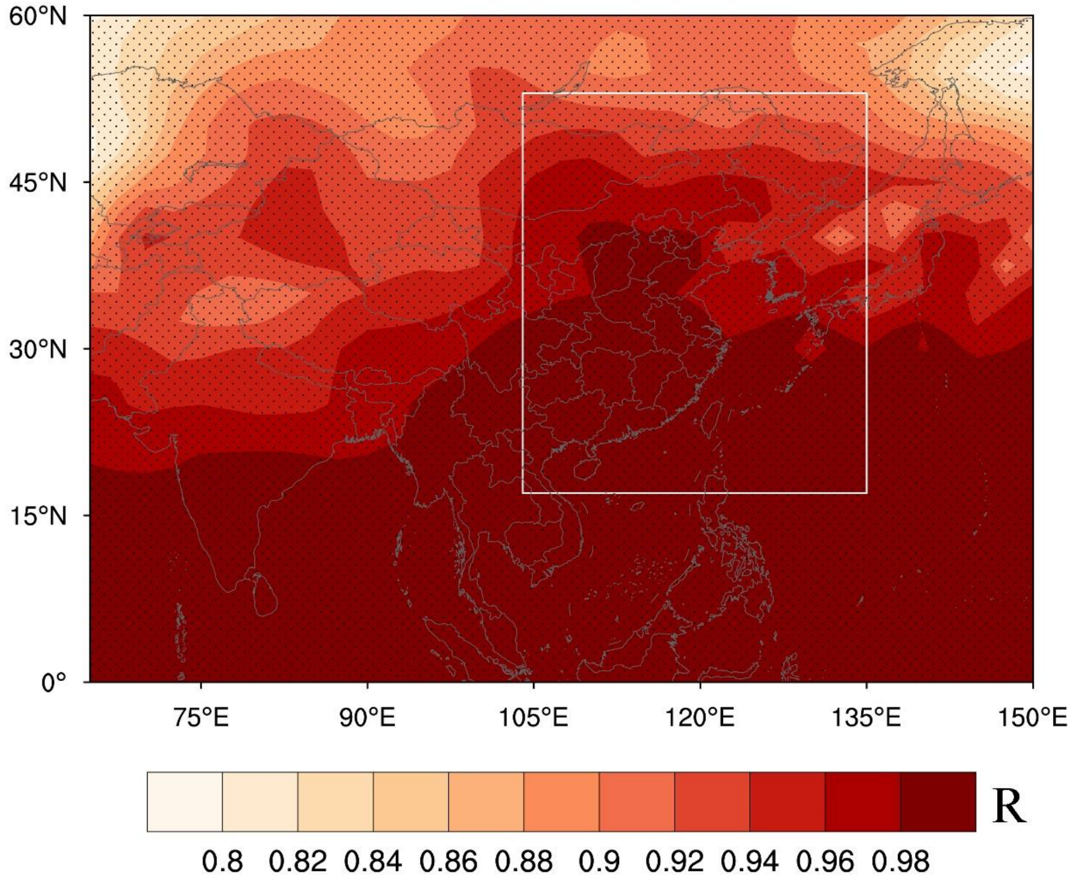

Fig. S3. The correlation of geopotential height between NCEP and ERA5 reanalysis data. The shading indicates the correlation, and the black dots indicate passing the 99% level of significance test.

Reference:

Guan, Q., Li, Q., SHI, C., HU, Y., MEI, C. and ZHANG, N.: Evaluation of Ｒeanalysis Data Based on the Three-dimensional High-density Sounding Data of the Qinghai-Tibet Plateau, Meteorol. Environ. Res., 12(1), 34-41+51, doi:10.19547/j.issn2152-3940.2021.01.007, 2021.

4. How many sounding profiles at 08, 14 and 20 BJT were used in this study for each studied city? How to use 08 and 20 LT soundings to estimate the afternoon BLH? Please clarify. In summer, the relationships between BLH and concurring/compound pollution in East China are quite complicated due to the transport of precursors (https://doi.org/10.1016/j.envpol.2020.115775). More in-depth analysis/discussion on the PBL-pollution linkage and transport of precursors in East China must be added.

RESPONSE: Thanks for your constructive suggestions. We have clarified this at lines 162–166 on pages 6-7 as follow: "Surface meteorological data, such as Tmax, precipitation, WS and RH from 611 meteorological observation stations, along with sounding data at 1400 Beijing time (BJT) from 64 stations and at 0800 BJT and 2000 BJT from 77 stations, in eastern China, were obtained from the China National Meteorological Information Center of the China Meteorological Administration (http://data.cma.cn/site/index.html).". The transportation of precursors and PBL-pollution linkage have been added in Discussion. Please also see as follow:

“In particular, Type 1 had significantly warmer temperatures over the boundary layer during the compound pollution periods of the BTH region, as compared with the clean periods. The daytime BLH under the compound pollution condition was also higher than that under the clean condition. In addition, there were different directions of prevailing winds during the two periods. The prevailing southerly winds during the compound pollution period may have driven the transportation of air pollutants from the southern plains, resulting in more serious pollution (Fig. 11; see also. Miao et al. (2020) also proposed another mechanism—that is, the synoptic southerly warm advections at the top of PBL, can strengthen the elevated thermal inversion layer and suppress the development of the PBL, causing worse pollution. Co-influenced by the topographical effect of the northern mountainous areas and the boundary layer structure, air pollutants could be trapped in the BTH region. In comparison, although there was a southerly prevailing wind in the BTH region (Figs. 11 and S14), the rain belt also being located in the southern area of the BTH might have led to the potential removal of $PM_{2.5}$ (Fig. 9j). Therefore, compound pollution across the BTH region might mainly have been due to local emissions of air pollutants”.

5. Please carefully check the cited papers, some were not properly. For example, the BLH estimation method was actually from the study of Seidel et al. (2012, https://doi.org/10.1029/2012JD018143).

RESPONSE: Sorry for our confusion. We have carefully checked the cited papers and revised in the latest manuscript. Thanks for your attention.

Anonymous Referee #2:

The authors use the T-mode PCA to objectively classify the summertime synoptic weather pattern across East-Asia and the western Pacific Basin aiming to identify the mode(s) most favorable for compound pollution events across sub-regions in China, specifically for PM2.5 and O3. Many factors governing these events operating across an array of scales are explored. The PCA identified 4 synoptic regimes characterizing the seasonal set up of the 500 hPa WPSH from 2015-2018. An additional large-scale circulation is also at work here, the East-Asian monsoon, which is discussed in context to the WPSH. Additionally, the authors discuss the effects of precipitation frequency and boundary layer characteristics on regulating compound pollution events. Occurrences of pollution are based on Chinese governmental standards.

The authors present a much-improved manuscript. The authors now show a clear connection between different synoptic modes and compound pollution events across different sub-regions in eastern Asia. The authors link the favorable synoptic modes to favorably meteorological conditions in Tmax, wind, stability, and more.

I believe that this paper will be ready for publication once its grammar has been improved. Thus, I recommend major revisions at this time, but I must emphasize that the authors should be proud of the improvements they have made to this manuscript. There is a strong message developing. With the proper grammatical improvements, this will be a significant contribution to the literature.

RESPONSE: Thank you very much for your high recognition of our work, and we believe that our revised manuscript will be further improved under your constructive suggestions. Please find our point-by-point responses below.

1. The abstract can and should be shortened considerably. The authors have identified two

preferred SWPs conducive for compound pollution events and then provide many details. The details can be left to the main text and omitted from the abstract. Furthermore, the abstract should be in the same tense. Currently, there is a mix of past tense and present tense expository.

RESPONSE: Thanks for your constructive suggestion. We have shortened the abstract and carefully checked the sentence tense, please also see as follow: "Surface ozone (O_3) pollution during summer (June–August) over eastern China has become more severe in recent years, resulting in a co-occurrence of surface O_3 and $PM_{2.5}$ (particulate matter with aerodynamic diameter ≤ 2.5 μm in the air) pollution. However, the mechanisms regarding how the synoptic circulation pattern might influence this compound pollution remain unclear. In this study, we applied the T-mode principal component analysis (T-PCA) method to objectively classify the occurrence of four synoptic weather patterns (SWPs) over eastern China, based on the geopotential heights at 500 hPa during summer (2015–2018). These four SWPs over eastern China were closely related to the western Pacific subtropical high (WPSH), exhibiting significant intraseasonal and interannual variations. Based on ground-level air quality observations, remarkable spatial and temporal disparities of surface O_3 and $PM_{2.5}$ pollution were also found under the four SWPs. In particular, there were two SWPs that were sensitive to compound pollution (Type 1 and Type 2). Type 1 was characterized by a stable WPSH ridge with its axis at about 22°N and the rain belt located in the south of the Yangtze River Delta (YRD); and Type 2 also exhibited WPSH dominance (ridge axis at ~25°N), but with the rain belt (over the YRD) at a higher latitude compared to Type 1. In general, SWPs have played an important role as driving factors of surface O_3–$PM_{2.5}$ compound pollution in a regional context. Our findings demonstrate the important role played by SWPs in driving regional surface O_3–$PM_{2.5}$ compound pollution, in addition to the large quantities of emissions, and may also provide insights into the regional co-occurring high levels of both $PM_{2.5}$ and O_3 via the effects of certain meteorological factors."

2. Line 79: What does "gradually been prominent" mean? Do the authors mean that O3 pollution in summer has increased in recent years?

RESPONSE: Indeed, the O_3 pollution in summer has increased in recent years. "For instance, Sun et al. (2016) showed that the observed summertime O_3 at Mt. Tai increased significantly by 1.7 ppbv yr^{-1} for the month of June and 2.1 ppbv yr^{-1} for the months of July–August during the period of 2003 to 2015. Furthermore, an increase in the maximum daily 8-h average concentration of O_3 (MDA8 O_3) at an annual-average rate of 4.6%, was reported by Fan et al. (2020), albeit with a decrease in the frequency of $PM_{2.5}$ pollution." are shown at lines 72–76 on pages 3.

References:

Fan, H., Zhao, C. and Yang, Y.: A comprehensive analysis of the spatio-temporal variation of urban air pollution in China during 2014–2018, Atmos. Environ., 220(November), 117066, doi:10.1016/j.atmosenv.2019.117066, 2020.

Sun, L., Xue, L., Wang, T., Gao, J., Ding, A., Cooper, O. R., Lin, M., Xu, P., Wang, Z., Wang, X., Wen, L., Zhu, Y., Chen, T., Yang, L., Wang, Y., Chen, J. and Wang, W.: Significant increase of summertime ozone at Mount Tai in Central Eastern China, Atmos. Chem. Phys., 16(16), 10637–10650, doi:10.5194/acp-16-10637-2016, 2016.

3. Line 90: WS, please define as wind speed. I do not it is defined previously in the main text.

RESPONSE: Sorry for our negligence, and thank you for your reminder. We have defined "WS" as wind speed at line 80 on page 4.

4. Line 105: When referring to previous studies, present material in the past tense, but the rest of the paper should be written in the present tense.
RESPONSE: Thanks for your kind suggestion. We have revised this sentence to "Miao et al. (2015) showed that RH was high when aerosol pollution occurred in the BTH region."

5. Line 116: "anomalies" should be "anomaly".
RESPONSE: Thanks, and revised.

6. Line 129: Are the winds southerly or northwesterly?
RESPONSE: The weak northwesterly prevailing winds was related to local emissions of aerosols, while the southerly prevailing winds was related to the transportation of pollutants from southern cities to Beijing.

7. Line 137: Delete "simulation"
RESPONSE: Deleted, and thanks.

8. Line 146: Should be "pollutants"
RESPONSE: Thanks, and revised.

9. Line 192: "consists" should be "consisting"
RESPONSE: Thanks, and revised.

10. Line 193: "pattern" should be "patterns"
RESPONSE: Thanks, and revised.

11. Lines 194-197: This sentence needs to be reworked grammar-wise.
RESPONSE: Thank you for your kind suggestion. We have revised it as follow: "First, the weather data are spatially standardized and split into 10 subsets by T-PCA. Then the principal components (PCs) of weather information are estimated by applying singular value decomposition, and the PC score for each subset can be calculated after oblique rotation. Finally, the resultant subset with the highest sum will be selected by comparing 10 subsets according to contingency tables, and its types can be output as well (Miao et al., 2017; Philipp et al., 2014).".

12. Line 205: Are the authors counting days as O3 and PM2.5 days when > 50% of the sites exceed the aforementioned thresholds? If so, the grammar here needs to be reworked.
RESPONSE: Thanks for your advice. We have rewritten this sentence as "In this study, we characterized regional pollution days as occurring when the average values of more than 50% of sites in this region exceeded the aforementioned thresholds.".

13. Line 231: "The" should be "the"

RESPONSE: Thanks, and revised.

14. Line 232: New sentence should begin at ", as a result"
RESPONSE: Thanks for your comments, and revised.

15. Lines 233-237: Are the authors referring to the total days in the 2015-2018 period?
RESPONSE: Sorry for our unclear statement. The results refer to the summer of 2015-2018. We have revised it as follow: "During the study period, the number of days of O3 pollution in the BTH, YRD, PRD, GZP, and NEM regions was 254, 133, 84, 165 and 96 respectively, while the number of days of PM2.5 pollution was only 93, 8, 0, 2 and 1, of which compound pollution occurred on 76, 7, 0, 2, and 0 days according to Chinese standards (the asterisks in Fig. 3 indicate the compound pollution events)."

16. Line 237: This sentence is repeated
RESPONSE: Thanks for your kind reminder. We have deleted this sentence in the revised manuscript.

17. Line 255: Wait – are Figs. 2-3 composited only on days characterized by SWPs 1-4? I thought these for all days? If for all days, delete "days for four SWPs"
RESPONSE: If I am not mistaken, it is line 245? Yes, all days during the research period was classified into SWPs 1-4. Thank you for your scrupulous review and sorry for our carelessness. This sentence should be "Based on this target, the number of pollution days for the five urban clusters were 194, 52, 16, 47, and 20, respectively (Fig. 3).".

18. Lines 247-248: Change the wording of this sentence." These results indicate that, despite PM2.5 reductions, compound pollution events deserve public attention." Delete the following sentence.
RESPONSE: Thanks for your constructive comment. And revised.

19. Lines 261-263: This sentence needs to be reworded from "which might…" onwards
RESPONSE: Thanks for your constructive comment. We have reworded this sentence as follow: "Low-level southerly monsoonal flow forming at the periphery of an anomalously enhanced WPSH, along with the transportation of warm and humid air from the ocean to East Asia, might also be responsible for the asymmetric spatial distribution of ground-level O_3 [i.e., a decrease in southern China but an increase over northern China (Zhao & Wang, 2017)]".

20. Line 273: Change "in" to "across"
RESPONSE: Thanks, and revised.

21. Lines 275-278: This sentence is hard to follow and needs to be reworked. For example, how can the sea-land interaction interact with the southeastern region across China? I think the authors can just explain the different spatial configurations of the different modes of the WPSH and leave discussion for later on when discussing the compound pollution event conditions.

RESPONSE: Thanks for your constructive comment. We have rewritten this sentence as "The southwest wind from the South China Sea might have combined with the southerly wind in the eastern periphery of the WPSH. As a result, southerly winds prevailed across southeastern China, while northern China was mainly controlled by the westerly trough.".

22. Line 535: "locating" should be located

RESPONSE: If I am not mistaken, it is line 355? Thanks, and revised.

23. Line 415: Prevailing…." winds?"

RESPONSE: Thanks for your kind reminder. We have added "winds" to after "prevailing".

24. Lines 418-420: Were the prevailing winds driving pollution transport from the southern plains? This sentence needs to be reworked grammatically.

RESPONSE: We appreciate your suggestions. Yes, they were. We have reworded it as "In addition, there were different directions of prevailing winds during the two periods. The prevailing southerly winds during the compound pollution period may have driven the transportation of air pollutants from the southern plains, resulting in more serious pollution (Fig. 11; see also Miao et al., 2019, 2020).".

25. Fig. 11: Panels are uneven. Please replot

RESPONSE: Thanks, and replotted.

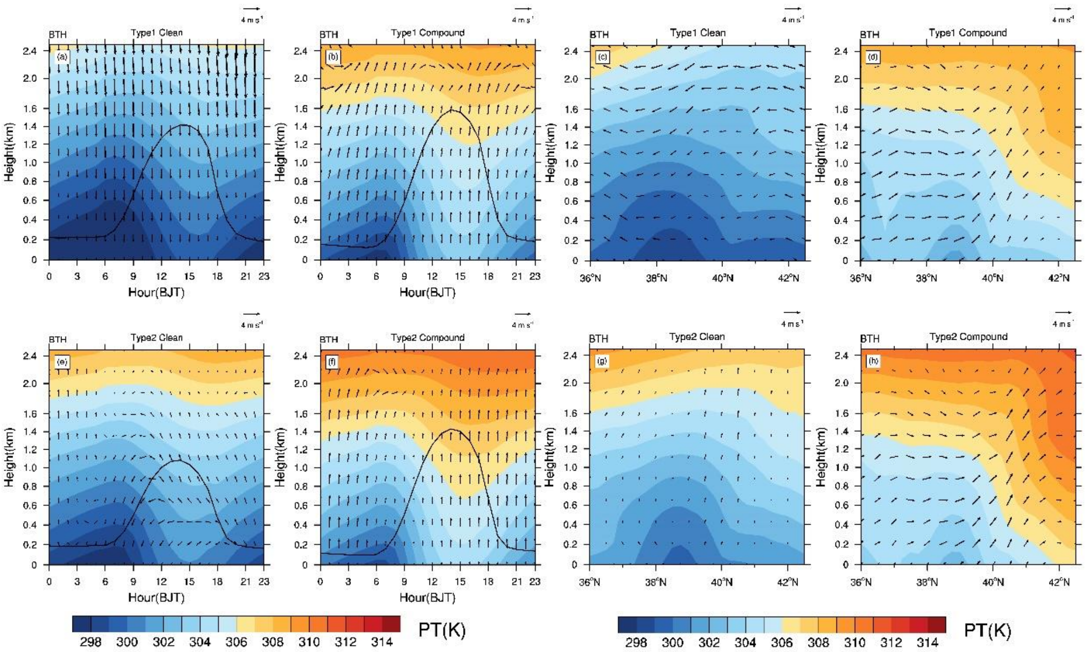

Fig. 11. Daily variations of horizontal wind, potential temperature and BLH in the BTH area during clean and compound pollution periods under Type 1 and Type 2 (a, b, e, f). The vertical cross-section of u-wind, w-wind and potential temperature for the same situation in the BTH region (c, d, g, h). The w-wind is multiplied by 100 when used. The data are from the ERA5 reanalysis.

26. Lines 427-463: These points can be shortened, and the grammar needs to be revised. A lot of

the discussion for this passage was made in previous sections.

RESPONSE: Thanks for your constructive suggestion. We have shortened and revised these points. Please also see as follow:

"(1) Type 1: Under the conditions of high temperatures (Tmax > 27°C), moderate humidity (RH ~60%), and low PF, photochemical reactions were greatly promoted to cause severe O_3 pollution. Meanwhile, the BTH–NYRD areas were located in front of the westerly trough, under the influence of the warm and humid air of the WPSH, and so the hygroscopic growth of fine particulates potentially caused a certain amount of $PM_{2.5}$ pollution (Li et al., 2017; Zhang et al., 2016b), becoming O_3–$PM_{2.5}$ compound pollution (Fig. 12). In addition, the prevailing southerly winds in the boundary layer were able to transport the pollutants emitted from southern cities to the BTH, atmospheric stratification was stable when the air mass was sinking (Miao et al., 2019b; Figs. 11 and S12), and compound pollution may have been especially severe. Although a relatively higher BLH occurred in the BTH region, the prevailing southerly winds in the boundary layer served to further increase the pollution.

(2) Type 2: O_3 pollution was severe under the meteorological conditions of high temperatures, moderate humidity, and weak precipitations. The $PM_{2.5}$ in the BTH region, which was located in front of the westerly trough, was high since the shallow boundary layer and low wind frequency were unfavorable for the diffusion of pollutants. Therefore, O_3–$PM_{2.5}$ compound pollution was also rather frequent (Fig. 12).

(3) Type 3: High temperatures, low humidity, and weak precipitations over the YRD region tended to generate a large amount of O_3, while the positive BLH and negative FLWD anomalies were unfavorable to O_3 accumulation. On the other hand, summer typhoon activities might have weakened the WPSH intensity over the YRD region, leading to the eastward retreat and northward shift of the WPSH. As a result, the high WS across coastal areas was able to ease the ground-level O_3 pollution (Shu et al., 2016). For the BTH and PRD regions, the high PF tended to suppress the production of O_3.

(4) Type 4: High temperatures, medium-high humidity and weak precipitations in the GZP and PRD regions were able to cause O_3–$PM_{2.5}$ compound pollution, but the $PM_{2.5}$ pollution in both regions was not heavy, possibly in relation to local lower emissions of pollutants. Under the control of the WPSH, there were strong photochemical reactions at high temperatures and little rainfall in some eastern regions (such as the northern BTH, YRD), which was also conducive to O_3 generation (Fig. 12). Meanwhile, relative to Type 1, O_3 pollution was lighter in the BTH, due to the differences of RH, BLH and FLWD.".