Dear editors and four reviewers:

Thank you all for your review and comments concerning our manuscript entitled “An important mechanism of regional O$_3$ transport for summer smog over the Yangtze River Delta in East China” (Manuscript ID: acp-2018-479). Those comments are all valuable and very helpful for revising and improving manuscript. We have studied comments carefully and have accordingly made the revisions. Revised parts are highlighted with Track Changes in the revised manuscript. In the following we quoted each review question in the square brackets and added our response after each paragraph.

For Referee #4:

Many thanks for your encouraging comments. We have revised the manuscript accordingly. All the revisions have been highlighted with Track Changes in the revised manuscript. The point-by-point responses to the reviewer’s comments are as follows:

**General comments:**

1. “The authors presented their efforts in applying observations and model simulation to analyze a severe O$_3$ pollution case in China. This is an important and interesting topic considering the adverse health effect of O$_3$. The materials are reasonably organized, and the unique horizontal transport and vertical mixing mechanisms were reported. Therefore I would recommend this manuscript to be published if the following concerns can be properly addressed.”

**Response 1:** We appreciate the reviewer’s positive comments on our manuscript. We have revised carefully the manuscript based on the following comments.

**Major comment:**

1. “Please consider rephrase the whole manuscript for English editing with help from native speaker. There are a lot grammar errors, confusing lengthy sentences, and improper wordings. Some are pointed out in the minor comments. The current shape is not acceptable for scientific journal publication.”

**Response 1:** Thanks for reviewer’s comments. We have rephrased the whole manuscript for English editing with the help of native speaker in modifying grammar errors, confusing lengthy sentences and improper wordings. Please find all the revisions highlighted with Track Changes in the revised manuscript.

2. “The unique transport and vertical mixing mechanism shall be discussed with more in-depth analysis, including:”

1) “first, background introduction is necessary to briefly describe the general condition of O$_3$ and meteorology over the study domain, thus the findings from examining the extreme event can be highlighted. For example, multi-year data of local O$_3$ observations (or satellite products) could be used to demonstrate the frequency, seasonality, and spatial distribution of O$_3$ smog in YRD. Climate data (e.g., observation from NCDC or China Meteorology Agency) could also be used to describe the general PBL condition in YRD;”
Response 1): Thanks for your suggestion. The according introduction has been added in the revised manuscript as follows (section 1 (paragraph 5)):

In recent years, ambient O$_3$ levels have enhanced over the Yangtze River Delta (YRD) in East China with more frequent pollution events from late May to July (Tang et al., 2013). During 1990 to 2013, the hourly O$_3$ peaks varied from 140 to 167 ppbv (about 294-350 μg m$^{-3}$) in the YRD region, from 160 to 180 ppbv (about 336-378 μg m$^{-3}$) in the Beijing-Tianjin-Hebei area over North China Plain and from 200 to 220 ppbv (about 420-462 μg m$^{-3}$) in the Pearl River Delta (Wang et al., 2017). Coupled with the increases of nitrogen oxides (NO$_x$) and volatile organic compounds (VOCs) emissions, O$_3$ distribution in the lower troposphere is significantly influenced by winds, air temperature, cloud cover, and downward shortwave radiation through changing the transport and chemical formation of O$_3$ (An et al., 2015; Gao et al., 2016; Xu et al., 2008; Li et al., 2018). O$_3$ levels could increase with a rate of 4–5 ppb K$^{-1}$ when temperature was between 28 and 38 °C (Pu et al., 2017). The prevailing winds driving transport of air pollutants from the YRD industrialized areas might have contributed to the O$_3$ enhancement (Tang et al., 2013). The ambient O$_3$ levels could be affected by the diurnal variation of atmospheric BL structure over YRD with nighttime stable BL height at 200 m and the daytime BL height reaching up to about 1200 m (Chang et al., 2016).

References:


2) “Second, discussion about the transport shall be improved with more solid demonstrations, many of the current statements were roughly made without sufficient proofs. For example, section 2 promotes
the hypothesis that the extreme O3 event was due to regional transport, yet no discussion was made to exclude the potential impact from local emission or photochemical production; "

Response 2): Thanks for reviewer’s comments. Following the comments, we have improved discussion about the transport in the revised manuscript as follows (sections 2.3 and 4.3):

Tropospheric O3 levels are controlled by regional transport of O3 and the precursors as well as photochemical production in closely associated with local emissions of O3 and its precursors. Considering weak changes of local emissions in short time, the WRF-Chem simulation with the hourly emissions of chemical species over YRD unchanged between August 24 and 25. To analyze the impact from photochemical production, we used the surface NO2 concentrations and total radiation irradiance (TRI) to analyze the change of photochemical production rates. There were no apparent changes of NO2 and TRI between August 24 and 25, indicating that photochemical production exerted less impact on the high O3 level on August 25 compared to regional O3 transport in nocturnal RL. The analysis of simulation results, revealed that vertical mixing from the upper O3-rich RL to daytime surface layer was a large contributor to O3 enhancement on August 25 (Fig. 7).

3) “Third, the most important one, the driving forces of the unique transport and mixing processes were not discussed at all. The authors spent a lot efforts to describe the severe O3 case and how it was accumulated though regional transport, but paid little attention to the causes. For example, in Fig.4(a), why O3 in 0-0.5km was depleted after 20:00, but remained high in 0.5-1km? The near surface layer NO might be responsible for titration but no demonstrate was made; Does the residual layer present in all seasons, and does it always host O3 or other atmospheric pollutants? The southeast wind shown in this study seems closely related with East Asia summer monsoon, thus does it also carry excessive O3 from the ocean into inland YRD? Fundamental questions such as what make the high O3 concentration in residual layer remained unsolved. These are the key findings that shall be reported in a journal publication.”

Response 3): Thanks for comments. Here are the responses for each of comments.

a) “Third, the most important one, the driving forces of the unique transport and mixing processes were not discussed at all. The authors spent a lot efforts to describe the severe O3 case and how it was accumulated though regional transport, but paid little attention to the causes.”

Response a): Thanks for comments. According to your suggestions, we have revised the manuscript as follows (sections 4.2 and 4.3):

Under the guidance of the prevailing easterly winds, the O3 transport from the eastern to western YRD region persisted during the nighttime from August 24 to 25, confirming our speculation about the regional O3 transport in the nocturnal RL over the YRD region (Figs. 4a and 4b). As the regional O3 transport reached the RL over the western YRD, O3 concentrations in the RL accumulated up to 200 µg m\(^{-3}\) over the western site NJ at 200 µg m\(^{-3}\) around 6 am in the sunrise hours of August 25. In accompany with the disappearance of the residual layer after sunrise, the vertical mixing initiated by convective and turbulent processes in the development of daytime convective boundary layer. The vertical mixing in the convective boundary layer after sunrise from the upper levels to the ground with the net downward transport flux reaching up to 40 µg m\(^{-3}\) h\(^{-1}\), contributing a considerable surface O3 accumulation to the O3 pollution during summer smog on August 25 in the western YRD region (Fig. 7).
b) “For example, in Fig. 4(a), why O3 in 0-0.5km was depleted after 20:00, but remained high in 0.5-1km? The near surface layer NO might be responsible for titration but no demonstrate was made;”

Response b): Based on the current understanding of atmospheric chemistry in boundary layer (BL), ambient O3 levels are strongly influenced by diurnal variation of the (BL) structure. The daytime BL, also known as the convective boundary layer (CBL), is directly affected by solar heating of the earth's surface. In the major part of CBL, which is the mixing layer (ML), air pollutant concentrations distribute nearly uniformly resulted from the convective turbulent mixing. The nocturnal BL is often characterized by a stable layer (SL) near the surface and an overlying residual layer (RL). The SL develops due to radiative cooling after sunset. Above the SL, the remnants of the daytime ML form the RL with initially uniformly mixed air pollutants remaining from the preceding daytime (Stull, 1988), and O3 is a representative remnant in the RL due to lack of the process of NO titration and dry deposition in nighttime (Xie et al., 2016; Sillman, 1999). Over the Eastern YRD, the SL at 0-0.5km and the RL at 0.5-1.0km were developed after 20:00 on August 24, leading to the O3 depletion in 0-0.5km and but remained high in 0.5-1 km overnight.

In section 4.3 (Fig. 7), we calculated the contribution rates of overnight chemical reactions to surface O3, which could represent the nocturnal consumption of NO titration with the negative contribution rates. During the nighttime from August 24 to 25, the average consumption of chemical reactions was about -8.0 µg m⁻³ h⁻¹, while it was -8.5 µg m⁻³ h⁻¹ over the preceding nighttime to August 24.

References:

c) “Does the residual layer present in all seasons, and does it always host O3 or other atmospheric pollutants? Fundamental questions such as what make the high O3 concentration in residual layer remained unsolved.”

Response c): Yes, the residual layer with could generally present in all seasons (Morris et al., 2010; Venzac et al., 2009; Neu et al., 1994) excepting the strong distribution of atmospheric circulation in free troposphere. The residual layer could host O3 or other atmospheric pollutants depending on changes of atmospheric pollutants and the interaction with atmospheric boundary layer.

Based on fundamental theory of atmospheric chemistry in boundary layer (BL), the daytime BL, also known as the convective boundary layer (CBL), is directly affected by solar heating of the earth's surface. In the major part of CBL, which is the mixing layer (ML), air pollutant concentrations distribute nearly uniformly resulted from the convective turbulent mixing. The nocturnal BL is often characterized by a stable layer (SL) near the surface and an overlying residual layer (RL). The SL develops due to radiative cooling after sunset. Above the SL, the remnants of the daytime ML form the
RL with initially uniformly mixed air pollutants remaining from the preceding daytime (Stull, 1988), and O$_3$ is a representative remnant in the RL due to lack of the process of NO titration and dry deposition in nighttime (Xie et al., 2016; Sillman, 1999).

References:


d) “The southeast wind shown in this study seems closely related with East Asia summer monsoon, thus does it also carry excessive O$_3$ from the ocean into inland YRD?”

Response d): We agree with the reviewer’s comments. The southeast winds, which are closely related with East Asian summer monsoon could carry excessive O$_3$ from the ocean into inland YRD. The influencing extension and strength of O$_3$ import from ocean to land could be a further study on air quality in YRD, especially for the coastal areas.

Minor comment:

1. “P2-L27: Spell “NO” before use it.”

Response 1: It has been done.

2. “P3-L64: change word “incomprehensively”

Response 2: We have changed it to “poorly”.

3. “P3-L68: This manuscript has no in-depth discussion of the “climate change of Asian summer monsoon” or its impact on O$_3$, I would suggest remove this sentence or add the related discussion”

Response 3: Thanks for suggestion. We have removed this sentence.

4. “A brief introduction of the typical O$_3$ concentration urban areas of China would be necessary, to clarify if the high O$_3$ in YRD is an area-dependent condition or a national wide issue.”

Response 4: Following the reviewer’s suggestion, we have added the discussion in manuscript as follows (Lines 71-77):

In recent years, ambient O$_3$ levels have enhanced over the Yangtze River Delta (YRD) in East China with more frequent pollution events from late May to July (Tang et al., 2013). During 1990 to
2013, the hourly O$_3$ peaks varied from 140 to 167 ppbv (about 294-350 μg m$^{-3}$) in the YRD region, from 160 to 180 ppbv (about 336-378 μg m$^{-3}$) in the Beijing-Tianjin-Hebei area over North China Plain and from 200 to 220 ppbv (about 420-462 μg m$^{-3}$) in the Pearl River Delta (Wang et al., 2017).

References:


5. “Table 1 & Figure 1: Are there multiple sites or is there only one site for each city? Please also provide the web source or reference for the observation data”

Response 5: The surface O$_3$ concentrations were averaged from multiple sites for each city, and meteorological variables were only one site for each city. We have provided the web sources for the observation data in the revised manuscript as follows (section 2.1):

The meteorological data were collected from China Meteorological Administration (http://www.cnemc.cn) and the air quality monitoring data from the national environmental monitoring network of China (http://www.mep.gov.cn). The meteorological data were observed at meteorological sites of each city (one site for each city), including wind speed (m s$^{-1}$) and direction (deg.) at 10 m above ground level, air temperature (°C) and relative humidity (%) at 2 m above ground level with a temporal resolution of 3 hours, and total radiation irradiance with a time resolution of 1 hour. The air quality monitoring data used in the paper were the mean value of multiple sites in each city, with the temporal resolution of 1 hour.

6. “P4-L85: Why wind speed is collected at 10m but temperature and relative humidity are collected at 2m? For evaluation purpose, WRF can output wind speed at both 10m & 2m, and NCDC has observation data for both too.”

Response 6: According to the World Meteorological Organization (WMO) standards, wind speed at 10 m and air temperature and relative humidity at 2 m are conventionally observed in the global weather monitoring network. Therefore, we have output wind speed and direction at 10 m, air temperature and relative humidity at 2 m to compare with observations.

Thanks for the information from reviewer. The NCDC observation data of meteorology could be used for further study.

7. “P4-L95-97: Do you try to compare Temperature & O$_3$ between western (NJ) and eastern YRD? Local emissions would be another factor determining O$_3$, the conclusion made in line#95-96 was made without solid demonstration.”

Response 7: We have compared statistics of air temperature and O$_3$ changes at 6 YRD sites based on the hourly observation data in Table 1. Local emissions would be another factor determining the ambient O$_3$ levels. However, there are usually less changes in local emissions from day to day. Therefore, we could exclude the impact of local emission change on the high O$_3$ level on August 25.
The conclusion made in line#95-96 has been revised as follows: It is generally accepted that high O$_3$ concentrations are accompanied by high air temperature with strong photochemical reactions (Filleul et al., 2006; Pu et al., 2017; Seinfeld and Pandis, 1986). Pu et al. (2017) found that O$_3$ level increases with a rate of 4-5 ppb K$^{-1}$ when temperature is between 28 and 38 °C in NJ of YRD (section 1 (paragraph 4)).

Table 1: Averages (Ave) of maximum 8-hour running mean surface O$_3$ concentrations ($\mu$g m$^{-3}$), mean air temperature (°C) over the periods of maximum 8-hour running mean surface O$_3$ concentrations and daily maximum air temperature (°C) during the heat wave episode with their standard deviations (Std) over August 22-25, 2016 at 6 YRD sites.

<table>
<thead>
<tr>
<th>Sites</th>
<th>Maximum 8-hour running mean O$_3$ Ave</th>
<th>Std</th>
<th>Mean air temperature Ave</th>
<th>Std</th>
<th>Daily maximum air temperature Ave</th>
<th>Std</th>
</tr>
</thead>
<tbody>
<tr>
<td>NJ</td>
<td>204.3</td>
<td>58.2</td>
<td>32.6</td>
<td>0.5</td>
<td>33.4</td>
<td>0.7</td>
</tr>
<tr>
<td>ZJ</td>
<td>163.3</td>
<td>44.7</td>
<td>32.6</td>
<td>0.7</td>
<td>33.2</td>
<td>0.8</td>
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<tr>
<td>CZ</td>
<td>174.4</td>
<td>34.8</td>
<td>33.2</td>
<td>0.7</td>
<td>34.2</td>
<td>0.9</td>
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<tr>
<td>WX</td>
<td>190.5</td>
<td>24.9</td>
<td>33.4</td>
<td>0.3</td>
<td>34.5</td>
<td>0.6</td>
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<tr>
<td>SZ</td>
<td>173.7</td>
<td>21.0</td>
<td>33.3</td>
<td>0.2</td>
<td>34.0</td>
<td>0.2</td>
</tr>
<tr>
<td>SH</td>
<td>141.1</td>
<td>7.8</td>
<td>32.0</td>
<td>0.4</td>
<td>32.7</td>
<td>0.3</td>
</tr>
</tbody>
</table>

References:

8. "P4-L95: ‘The O$_3$ concentrations over NJ of the western YRD were much higher...’ this is not professional scientific writing, please describe it with exact numbers."

Response 8: Thanks for suggestion. We have revised as follows (section 2.2):
The O$_3$ concentrations over NJ of the western YRD were 10-63 μg m$^{-3}$ higher than the eastern YRD region (CZ, WX, SZ and SH) during this heat wave episode.

9. "P5-L99: ‘Surface air temperature and solar radiation, deeply affect photochemical production.’ Please rewrite this sentence or remove it, these are unnecessary common sense for journal publication."
Response 9: It has been removed.
10. “P5-L101: ‘exhibited’ shall be ‘showed’?”
Response 10: It has been revised.

11. “P5-L102-105: Please rewrite this lengthy sentence, either break it into a few short ones or rephrase.”
Response 11: It has been rewritten as follows (section 2.3 (paragraph 1)):

The maximum 8-hour running mean O_3 concentrations increased from 230.1 μg m\(^{-3}\) on August 24 to 284.8 μg m\(^{-3}\) on August 25 2016 and maximum hourly O_3 concentrations enhanced from 256.8 μg m\(^{-3}\) on August 24 to 317.2 μg m\(^{-3}\) on August 25 2016, presenting an obvious enhancement in western urban site NJ. In contrast, surface maximum total radiation irradiances and maximum air temperature respectively decreased from 896 W m\(^{-2}\) and 34.1 °C to 872 W m\(^{-2}\) and 33.9 °C during the two days.

12. “P5-L103: ‘NJ of the western YRD’ this term has been used several times in the manuscript, I would recommend simply using ‘NJ’ or ‘the western part of YRD’.”
Response 12: We have changed it to “NJ” there.

13. “Fig.2 & Table2: Why the data from other sites were not shown?”
Response 13: We have showed the statistics on the observational data of all the YRD sites in Table 1. Following review’s suggestion, here we present the hourly changes of O_3, air temperature, wind speed and direction in six cities in the following Figures S1, S2 and S3.

![Fig. S1 Hourly series of surface O_3 concentrations in NJ, ZJ, CZ, WX, SZ and SH.](image)
Fig. S2 Time series of 2 m air temperature in NJ, ZJ, CZ, WX, SZ and SH.

Fig. S3 Time series of 10 m wind speed and direction in NJ, ZJ, CZ, WX, SZ and SH.
14. “P5-L108: Unnecessary, in addition to local production and transport, what else can result in high O3?”
Response 14: In many studies, local production and transport of O3 are the major sources to high O3.

15. “P5-L110: ‘it is estimated that the daily mean surface NO2 concentrations varied slightly during August 24 and 25’. Analysis of NO2 is important and necessary to be included as it supports your conclusion.”
Response 15: Thanks for your comments. We agree with the comments. Analysis of NO2 is important and necessary to be included as it supports your conclusion.

16. “P6-L130: Latest MEIC updates the emission to 2015, if the 2012 emission was not projected to 2016, it’s better to rerun the simulation with latest emission inputs.”
Response 16: Following the reviewer’s suggestion, we have rerun the simulation with the latest MEIC emission inventories of 2015 and analyzed the updated simulation over YRD in the revised manuscript, although there are small differences of O3 simulation over the YRD region between MEIC emissions 2012 and 2015.

17. “P6-L134: Incorrect grammar, it shall be ‘Simulated wind speed, air temperature, relative humidity, and O3 concentrations are compared with observations...’”
Response 17: We have revised as your suggestion as follows (section 3.2):
Simulated wind speed, air temperature, relative humidity, and O3 concentrations are compared with observations at six sites over the YRD (Fig. 1b) during August 22-25, 2016 for the O3 pollution episode (Fig. 3).

18. “Section3.2: More evaluation statistics, such as normalized mean bias and root mean square error shall be applied to demonstrate model performance. Fig.3 cannot tell the absolute values of simulation bias. P6-L120-125 listed details of model configuration but no reason was given to clarify why these options were selected. It’s also necessary to briefly compare the simulation performance with other published WRF-Chem applications over YRD region.”
Response 18: Thanks reviewer’s suggestion. We have calculated these statistics in Table. S1. The simulation reasonably captures the observed changes of O3 and meteorology during the summer smog episode over the YRD.

We have briefly compared with the previous WRF-Chem studies over YRD region (Table S2), to optimize the simulation configurations in our study, and the simulation result in manuscript had a good performance compared those studies’ result.

Table S1 MB, NMB and RMSE of O3 (μg m^{-3}), wind speed (m s^{-1}), temperature (°C) and relative humidity (%) between simulation and observations.

<table>
<thead>
<tr>
<th></th>
<th>O3</th>
<th>Wind Speed</th>
<th>Temperature</th>
<th>Relative Humidity</th>
</tr>
</thead>
<tbody>
<tr>
<td>NJ</td>
<td>MB</td>
<td>-18.7</td>
<td>0.7</td>
<td>-0.1</td>
</tr>
<tr>
<td></td>
<td>NMB</td>
<td>-17.6%</td>
<td>25.5%</td>
<td>-4.3%</td>
</tr>
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</table>
RMSE  36.8  1.3  1.5  9.7
ZJ
RMSE  29.5  2.6  1.7  8.7
NMB  -12.8%  108.7%  -2.8%  0.7%
ZJ
RMSE  30.1  1.6  2.9  11.2
NMB  -12.6%  47.9%  -8.2%  10.0%
CZ
RMSE  37.2  1.6  2.8  13.7
NMB  -22.1%  47.0%  -8.6%  18.0%
WX
RMSE  34.6  1.7  3.0  14.6
NMB  -17.9%  59.0%  -9.2%  19.5%
SZ
RMSE  31.9  2.6  1.7  9.7
NMB  -21.9%  68.0%  -4.3%  12.7%
SH

MB (mean bias) = \( \frac{\sum_{i=1}^{n}(S_i - O_i)}{n} \)
NMB (normalized mean bias) = \( \frac{\sum_{i=1}^{n}(S_i - O_i)}{\sum_{i=1}^{n} O_i} \ast 100\%
RMSE (root mean square error) = \( \sqrt{\frac{\sum_{i=1}^{n}(S_i - O_i)^2}{n}} \)

Where Si and Oi are the simulated and observed value.

Table S2 Model configuration in some other references.

<table>
<thead>
<tr>
<th>Item</th>
<th>Our work</th>
<th>Gao et al</th>
<th>Zhang</th>
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</table>

References


Response 19: We have changed it to “Analysis of”.

21. “P7-L153: It’s necessary to include a brief introduction of the climatology in NJ area before using “heat wave”.”
Response 20: We have revised and added introduction of climatology in YRD and NJ in manuscript as follows (section 2.2):

Due to the western Pacific subtropical high staying over YRD region in summer, it always leads to high temperature and heat wave. The average of daily temperature and daily maximum temperature in YRD were 27.1 and 39.5 °C in summer during 2013-2016, and the average of summer daily temperature was 28.5 °C in NJ.

22. “Fig.4: Need a clear definition of ‘eastern’ and ‘western’ if you are showing subdomain averages in the figure.”
Response 21: In the manuscript, the western YRD covered the site NJ, and the eastern YRD included CZ, WX, SZ and SH.

23. “P7-L165: Please rewrite this lengthy and confusing sentence.”
Response 22: We have revised these (section 4.1 (paragraph 4)):

We compared the temporal changes of O3 “reservoir” in the nocturnal RL over the western and eastern YRD areas in Figures 4a and 4b. It is interesting that the eastern O3 “reservoir” obviously leaked with reducing the O3 concentrations over the nighttime of August 24 (Fig. 4a), while the western O3 “reservoir” was gradually strengthened, forming a high O3 center exceeding 200 μg m⁻³ around 6 am on August 25.

24. “P8-L175: Please change the word “questionable”, check it in the dictionary before using it.”
Response 23: We have changed it to “worth discussing”.

25. “Fig.5: No prominent changes of O3 or wind stream are shown, why use 4 subpanels?”
Response 24: Fig. 5 shows the growth of cyclone circulation over NJ from 00:00 to 09:00. The center of cyclone circulation moved from southwest to south of NJ, and the wind direction changed from east-southeast to southeast at 900 m in residual layer. Hence, due to the cyclone circulation over NJ from 06:00 until 09:00, high O3 converged in the RL over NJ and ZJ at the sunrise (Fig. 5).
26. “Fig. 6 cross sections are drawn along the red line in Fig. 1. If the observation along this track is not discussed, I would recommend to make cross-sectional figures along the travel path in Fig. 5.”

Response 25: Fig. 5 is focused at cyclone circulation over NJ and ZJ in western YRD from 00:00 to 09:00 on August 25. Fig. 1 shows the complete YRD region containing both western and eastern YRD region. Since the cross section in Fig. 6 shows the full scope from east to west during August 24-25, it is better to use the red line in Fig. 1 to show the travel path.

27. “P9-L201: Please specify how “vertical mixing” is calculated, if it is directly output by WRF-Chem, a bar chart would be better for Fig. 7 to present the contributions from all processes.”

Response 26: Thanks for comments. The contribution from “vertical mixing” is output by WRF-Chem. Actually, we have tried to plot a bar chart for Fig. 7, and the line-symbol figure was better to express temporal changes.