Reply for Anonymous Referee #2

Reviewer's comments on the paper by Ding et al. entitled "Year-round record of nearsurface ozone and "O_3 enhancement events" (OEEs) at Dome A, East Antarctica" submitted to Atmospheric Chemistry and Physics

The manuscript is within the scope of ACP. It presents scientifically significant material based on surface ozone measurements at three Antarctic stations. Of especial importance are data of measurements at Dome Argus, the highest Antarctic plateau (∽ 4000 m above sea level). However I have a lot of comments to the manuscript, which are listed below. The manuscript needs major revision.

Total response: Thank you for your comments and suggestions on our article. We revised and explained the errors and insufficient explanations in the article one by one according to the suggestions. The article has undergone great changes: (1) we have made great amendments to the contents and illustrations of section 3, adding references and deleting subjective judgments. (2) We have greatly revised the content of Section 4.2. We have re-analyzed this section by re-determining the weight of PSCF. (3) We made a major revision to section 4.3, contacted Dr. Putero, and cooperated in STEFLUX analysis. The conclusion of the article has been corrected accordingly.

Thank you for your constructive and insightful criticism and advice. We addressed all the points raised by the reviewer as summarized below.

1. One significant disadvantage of the manuscript is that some explanations of analysis results look like mere assertions. They are specified in more detail in the specific comments section.

Reply: The new manuscript has been checked carefully and we think these assertions has been improved.

2. The authors repeatedly expressed about importance of photochemical source of near-surface ozone in the Antarctic without providing evidence of it. Presumably they do not have clear idea of photochemical production of tropospheric ozone. See especially page 10.

Reply:According to your suggestion, this part has been modified and replaced by new analysis and description. Please find in the Line 255~269.

3. Inconsistent scientific language is often used in the manuscript. English should be generally improved.

Reply: The full text has been retouched and edited, and the English language has been

improved.

4. The potential source contribution function (PSCF) is corrected by multiplying it by some weights suggested earlier by other authors. However these weights are arbitrary and do not have any physical or mathematical reason. They modify arbitrary the distribution of the PSCF but do not allow estimating its statistical significance. I suppose that analysis of the PSCF distribution has to be done with accounting for statistical significance. Estimating statistical significance should take into account the fact that close-in-time trajectories are not independent. Without knowing whether the PSCF distribution is statistically significant one cannot rely on Fig. 6. Perhaps the following paper will help: Shikurov and Shukurova, Source regions of ammonium nitrate, ammonium sulfate, and natural silicates in the surface aerosols of Moscow oblast, Izvestiya, Atmos. Oceanic Phys. 2017, v. 53, p. 316-325, doi: 10.1134/S0001433817030136.

Reply: According to your suggestion, we have performed new experiments to better clarify the PSCF. We first cluster all the air mass trajectories during OEE and NOEE. Then we recalculate the track weights of different periods. Through recalculation of PSCF, we find that there are obvious differences in the potential pollution sources in different periods. The results are in good agreement with those of cluster analysis. The new weight calculation and graph are shown below.

$$W_{ij(NOEE)} = \begin{cases} 1.00 & nij > 12Nave \\ 0.70 & 12Nave > nij > 3Nave \\ 0.42 & 3Nave > nij > 1.5Nave \\ 0.05 & Nave > nij \end{cases}$$

$$W_{ij(OEE)} = \begin{cases} 1.00 & nij > 8Nave \\ 0.70 & 8Nave > nij > 2Nave \\ 0.42 & 2Nave > nij > 1Nave \\ 0.05 & Nave > nij \end{cases}$$

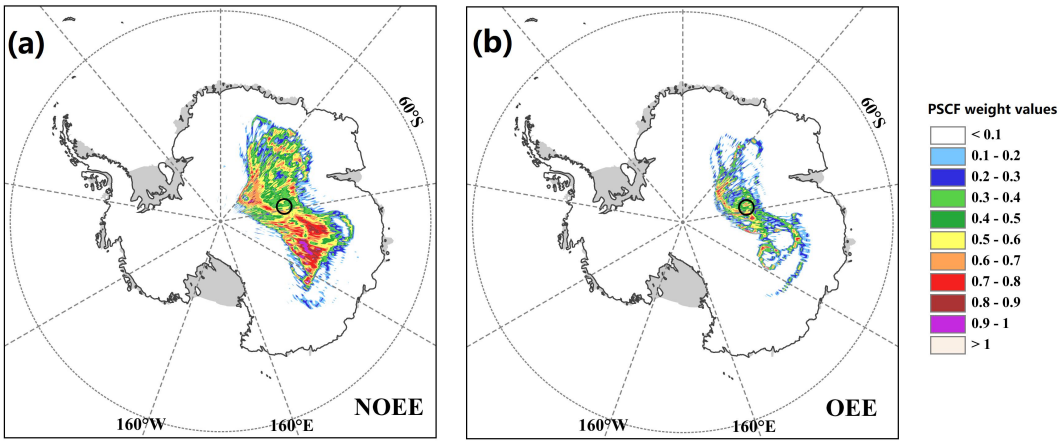
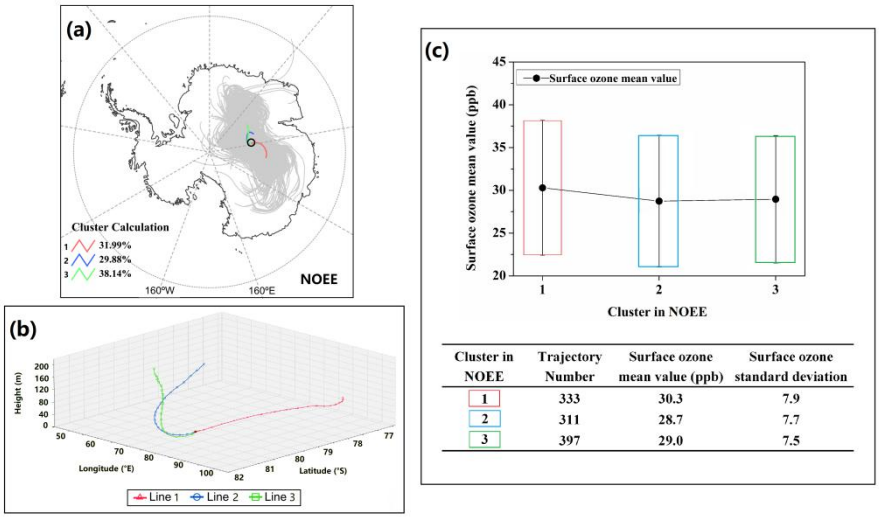
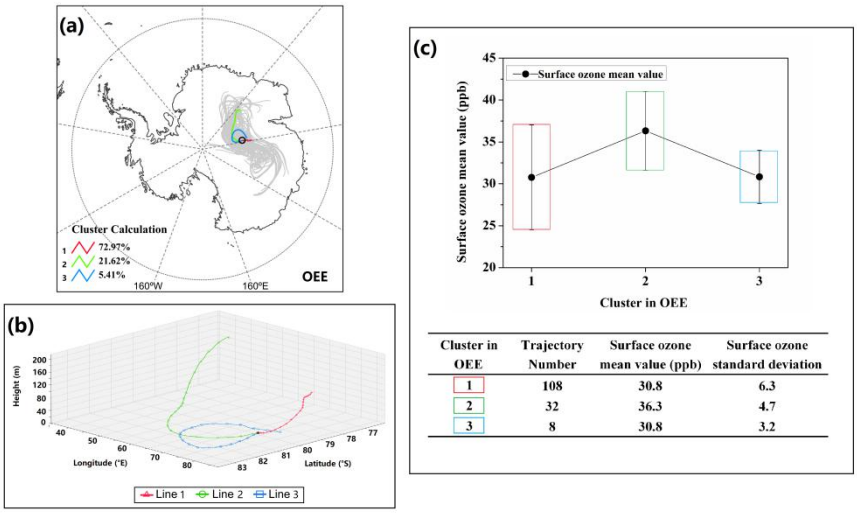


Figure 7. Likely source areas of surface ozone at Kunlun Station during the NOEE (a) and OEE (b) identified using PSCF (Potential Source Contribution Function).



Cluster in NOEE	Trajectory Number	Surface ozone mean value (ppb)	Surface ozone standard deviation
1	333	30.3	7.9
2	311	28.7	7.7
3	397	29.0	7.5

Figure 8. Backward HYSPLIT trajectories for each measurement day (gray lines in the Fig.8a), and mean back trajectory for 3 HYSPLIT clusters (colored lines in the Fig.8a, 3D view shown in the Fig.8b) arriving at Kunlun Station during the NOEE. Mean trajectory of 3 HYSPLIT clusters arriving at Kunlun Station during the NOEE. Subplot (c) shows the range of surface ozone mixing ratios measured at Kunlun Station by cluster.



Cluster in OEE	Trajectory Number	Surface ozone mean value (ppb)	Surface ozone standard deviation
1	108	30.8	6.3
2	32	36.3	4.7
3	8	30.8	3.2

Figure 9. Same as Fig. 8, but for OEEs.

5. Potential vorticity (PV) in the southern hemisphere polar stratosphere is generally negative. However values of PV in Fig. 7 are of inverse sign. This contradicts also to PV distribution in Fig. 10.

Reply: We have made great changes to the content of the fourth section of the article, and used the STEFLUX tool to analyze STT events. In combination with your suggestion, we have corrected the error.

6. Values of ozone concentration are given with excessive accuracy. One decimal place is enough.

Reply:According to your suggestion. The ozone concentration value in this paper is accurate to one decimal place.

7. There are no references to Fig. 4 and Fig. 7e in the text.

Reply:According to your suggestion, this part has been modified and replaced by new analysis and description. please find in the Line 257 and Line286.

8.Some works that are referenced to in the text are absent in the reference list.

Specific comments

L45-46. Add reference(s) to confirm this.

Reply: The original text is changed to: ozone (O_3) photochemical production in the troposphere occurs by hydroxyl radical oxidation of carbon monoxides (CO), metal (CH_4) and non metal hydrocarbons (generally referred to as NMHC) in the presence of nitrogen oxides (NOx) (Monks et al., 2015).

L61-62. Add reference(s) regarding the depth of the mixing layer.

Reply: According to your suggestion, we have added reference(s) regarding the depth of the mixing layer. Please find in the Line 67.

Berman S., Ku J Y., Rao S T.: Spatial and Temporal Variation in the Mixing Depth over the Northeastern United States during the Summer of 1995, Journal of Applied Meteorology, 38(12):1661-1673, https://doi.org/10.1175/1520-0450(1999)038<1661:SATVIT>2.0.CO;2, 1999.

L73-76. The downward transport of ozone is important not only on high-altitude terrains. Note also that stratospheric ozone in the polar regions can be transported to the troposphere not only during intrusion events but also as a result of slow but prolonged subsidence. In this sentence, references would be more appropriate to papers concerning polar regions (e.g., Gruzdev and Sitnov 1993; Roscoe 2004, Possible descent across the "Tropopause" in Antarctic winter, Adv. Space Res., v. 33, p. 1048- 1052; Greenslade et al. 2017, Stratospheric ozone intrusion events and their impacts on tropospheric ozone in the Southern Hemisphere, Atmos. Chem. Phys. V. 17, p. 10269-10290).

Reply: According to your suggestion, I have made changes and added explanations. The original text is changed to: "The near surface ozone concentrations at high-elevation sites can also be increased by the downward transport of ozone rich air from the stratosphere during deep conversion and stratosphere to troposphere transport (STT) events. More, the stratospheric ozone in the polar regions can be transported to the troposphere not only during intrusion events but also as a result of slow but prolonged subsidence. (e.g., Gruzdev et al., 1993; Roscoe et al., 2004; Greenslade et al. 2017,)."

L91-92. Unclear. Why does it lead to overestimation?

Reply: The words used here are not accurate. We changed "overestimation" to "inaccurate estimation".Please find in the Line 104.

L125-126. Specify address.

Reply: The Amundsen-Scott Station (89° 59'51.19 "S, 139° 16'22.41" E, altitude 2835 m) is located at the SP and operated by the United States. The near-surface ozone data were downloaded from the Earth System Research Laboratory Global Monitoring Division under the NOAA (https://www.esrl.noaa.gov/gmd/dv/data).

L154. What is PM?

Reply: The words used here are not accurate. The original text is changed here to: “If the total number of end points that fall in the cell is nij and there are mij points for which the measured ozone parameter exceeds a criterion value selected for this parameter, then the conditional probability, the PSCF, can then be defined as:”

L180-184. This paragraph is somewhat misleading. It reduces the ozone annual variation to change between polar day and polar night. However Fig. 2 shows that large values of ozone concentration peculiar to polar night are also observed for long time intervals before or/and after the polar night period. Similarly, low ozone concentration values peculiar to polar day are observed after the polar day period.

Reply: Yes, the comparison of the concentration characteristics of polar day and night is mainly in Figure 3.

L185. Wrong statement. According to Fig. 2, Ozone concentration at the SP during polar night is generally less than at the Kunlun station.

Reply: Yes, there is a mistake here.The SP had the highest near-surface ozone concentration during non-polar night. The average concentration during this period was 28.1 ppb.

L191-192. Gruzdev et al. -> Gruzdev and Sitnov. Oltmans et al. 1976 and Ghude et al. 2010 are absent in the reference list. Probably you mean Oltmans and Komhyr 1976, Surface ozone in Antarctica, JGR, v. 81, p. 5359-5364.

Reply:There is a mistake in citation. Thank you for your correction. The revised version has been amended.

Oltmans, S. J Komhyr, W. D.: Surface ozone in Antarctica, Journal of Geophysical Research, v. 81, p. 5359-5364. http://dx.doi.org/10.1029/jc081i030p05359,1976.

L193-196. Unfounded statements. Please confirm these by references or give clear arguments.

Reply:In Antarctica, the emissions of ozone precursors are generally less than those at mid and low latitudes, whereas ultraviolet radiation is relatively strong; thus, When there is solar radiation,the depletion effect is much greater than the effects from photochemical reactions

during the warm season (Schnell et al., 1991).

L198-199. Unreasonable explanation. Why weaker variability is due to short polar night?

Reply: During the polar night, due to the lack of light, photochemical reaction stopped. And Due to the lack of loss effect, the ozone concentration gradually increases and the fluctuation becomes smaller. During the polar night, the monthly variation of surface ozone in ZS is smaller, higher than SP, but lower than DA.

L200-201. This explanation is not sufficiently reasoned since it refers to literature sources one of which is absent in the reference list and the other is an abstract.

Reply: However, due to strong UV radiation in low latitude areas and the presence of bromine controlled ozone depletion events in coastal areas, the station shows a large seasonal variation during the non polar night (Wang et al., 2011; Prados-Roman et al., 2017).

L204-205. This explanation is not sufficiently reasoned since it does not follow from the references given.

Reply: This part is reinterpreted. The original part has been deleted.

However, at the Amundsen-Scott Station, the largest standard deviation was observed in December, similar to the characteristics at DC from November to December (Legrand et al., 2009; Cristofanelli et al., 2018). Figures 3 and 4 show that the near-surface ozone showed obviously larger variations at DA than at the SP during the polar night.Due to the difference of geographical location, the meteorological conditions of DA and SP are different. The abnormal fluctuation of ozone concentration over DA during the polar night may be related to its special geographical environment.

L205-206. Misconception. Enhanced variability does not require a special ozone source.

Reply: The original part has been deleted.

However, at the Amundsen-Scott Station, the largest standard deviation was observed in December, similar to the characteristics at DC from November to December (Legrand et al., 2009; Cristofanelli et al., 2018). Figures 2 and 3 show that the near-surface ozone showed obviously larger variations at DA than at the SP during the polar night.Due to the difference of geographical location, the meteorological conditions of DA and SP are different. The abnormal fluctuation of ozone concentration over DA during the polar night may be related

to its special geographical environment.

L207-209. The explanation is unfounded.

Reply: The original part has been deleted.

L231-232. Papers by Oltmans et al. 1976 are absent in the reference list (see comment to L191-192). Gruzdev et al. is also absent in the reference list. However it is relevant and can be added: Gruzdev, Elokhov, Makarov and Mokhov, 1993, Some recent results of Russian measurements of surface ozone in Antarctica. A meteorological interpretation, Tellus, v. 45B, p. 99-105.

Reply: Thanks for your suggestion, this error has been corrected. And added references and explanations.

L219-234. It would be relevant to refer to Fig. 4 here. One interesting feature in Fig. 4 is the presence of a specific and very regular diurnal variation at the DA station during the polar day period. You could try to associate it with the slope katabatic winds which have diurnal cycle in summer (see aforementioned reference to Gruzdev et al. 1993). Although these winds are most prominent off the plateau they, due to their large horizontal scale, can induce slow subsidence of the air in the boundary layer over plateau and therefore influence the surface ozone concentration because of vertical ozone gradient.

Reply: Thanks for your suggestion. The section 3.3 has changed a lot. The error has been changed.

To characterize the typical monthly O3 diurnal variations at these three stations, we analysed the mean diurnal variations of O3 at the three stations (Figure 4) and the standard deviation of the mean diurnal variations (Figure 5). At the DA site, the mean diurnal concentrations of each month were relatively steady, and with the standard deviation of the mean diurnal concentration of each month was lower than 0.4 ppb. At the SP, the mean diurnal concentrations were less variable as well. Except for December, the standard deviation of the mean diurnal concentration was lower than 0.3 ppb. At the ZS, except for October, the standard deviation of the mean diurnal concentration is greater than that in the other two stations. In particular, the standard deviation of the mean diurnal concentration of the ZS in September, November and December exceeded 0.5 ppb. The mean diurnal variations in different time periods were not obvious, and the mean diurnal concentrations of the three stations fluctuated within a range of less than 1 ppb, indicating that daily photochemistry reactions did not have the main impacts on the overall characteristics of near-surface ozone at the three stations. The magnitude of the diurnal variation was low, which is similar to the

variations found at other Antarctic stations (Gruzdev et al., 1993; Ghude et al., 2005; Oltmans et al., 2008).

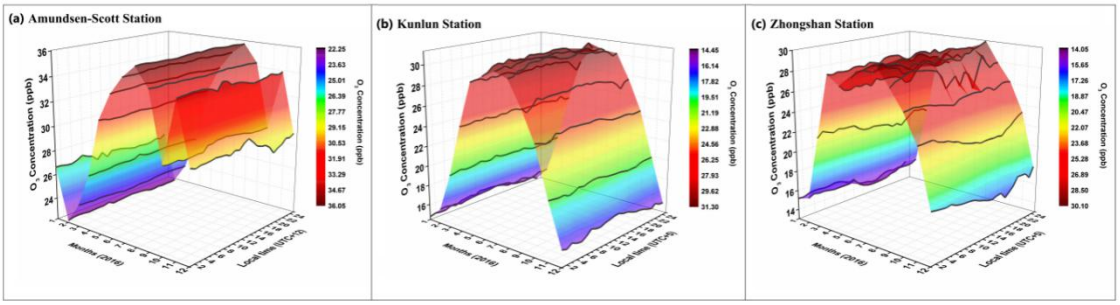


Figure 4. Mean diurnal variations in near-surface ozone concentrations at the Amundsen-Scott Station (a), Kunlun Station (b) and Zhongshan Station (c) during 2016

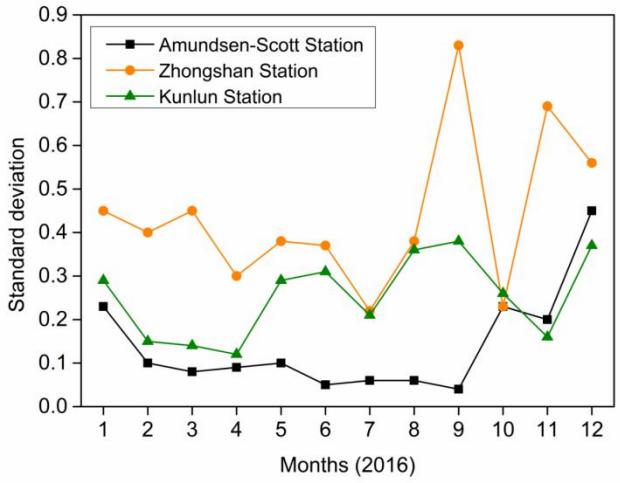


Figure 5. The Standard deviation of mean diurnal variations in near-surface ozone concentrations at the Amundsen-Scott Station, Kunlun Station and Zhongshan Station during 2016

L241-249. This part should be revised or removed.

Reply: Thanks for your suggestion. The section 3.3 has changed a lot. This part has been removed.

L241-242. Are hydrocarbons really produced in surface snow?

Reply: There is a mistake in the expression here. The photodegradation on the snow surface only releases NOx and does not contain hydrocarbons. The section 3.3 has changed a lot. This part has been removed.

L243. Wrong reaction.

Reply: Thanks for your suggestion, this error has been corrected. The section 3.3 has changed a lot.

L245. What do you mean by a chain reaction?

Reply: The section 3.3 has changed a lot. The error has been changed.

L245-246. Inconsistency: production occurs when loss (destruction) occurs.

Reply: The section 3.3 has changed a lot. The error has been changed.

L248-249. Why the cold is the reason of the variation?

Reply: The section 3.3 has changed a lot. The error has been changed.

L259. What is meant by a well-mixed state? Does it have to do with atmospheric mixing?

Reply: Here is a hypothesis, if the Gaussian distribution is consistent, it represents an idealized good mixing state of the atmosphere.

L258-265. This procedure is not completely clear and internally contradictory. First, it is hypothesized that data falling out of the Gaussian distribution are "abnormal". But then the Gaussian fit is applied to these data.

Reply: Thanks for your suggestion, this error has been corrected. We have reinterpreted and modified this part.

The algorithm proposed in Cristofanelli et al. The analysis of OEEs was restricted to years 2016. Our method to select the days characterized by OEEs is based on a two-steps procedure. The first step consists in fitting the annual cycle of O3 mean daily values with a sinusoidal curve. This represents an "undisturbed" O3 annual cycle, not affected by the occurrence of summer O3 events. In the second step, the probability density function (PDF) of the deviations from the sinusoidal fit is computed, considering all of the daily data. Then, a Gaussian fit was applied to the obtained PDF. As reported by Giostra et al. (2011), the Gaussian distribution corresponds to a well-mixed state, given the hypothesis that instrumental errors and natural background variability follow a Gaussian distribution. The

deviations from the Gaussian distribution (calculated by the Origin 9© statistical tool) would indicate observations affected by non-background variability. To obtain a threshold value for selecting non-background O3 daily values possibly affected by "anomalous" O3 enhancements, we calculated a further Gaussian fit for the PDF points falling above 1 σ (standard deviation) of the Gaussian PDF, and we considered the intersection between the two curves as our threshold value (i.e., 3.4 ppb at the SP, 3.4 ppb at DA, 2.5 ppb at the ZS).Figure 6a, 6b, 6c shows the OEEs and "NO O3 enhancement events" (NOEEs) at these three stations, while Figure 6d, 6e, 6f reports the distribution frequency of OEE.

L267 and further. Two significant digits are enough.

Reply: Some of them are reduced.

L285. Do you mean the time or spatial scale?

Reply: In this paper, we consider the spatial scale.

L296. Air mass circulation? What is it? In meteorology, air mass is a volume of air which covers many hundreds or thousands kilometers in horizontal direction and hundreds meters or a few kilometers in vertical direction.

Reply: The error has been changed. Air mass circulation ->Air mass transport

L297-309. See general comment 4. It is very probable that at least part of the PSCF is statistically insignificant. From my point of view, the main conclusion from the back trajectory analysis is that all the 5-day trajectories depicted in Figs 6a, b are located around the plateau and do not have their origin out of the continent.

Reply: Thanks for your suggestion. In the new section 4.2, we analyze the potential sources of the backward trajectory during NOEE and OEE respectively. In this way, we can clearly see the difference between the potential sources of two different periods.

L310. Simulated? Did you do your own simulations or use HYSPLIT?

Reply: The section 4.2 has changed a lot. The error has been changed. We used HYSPLIT to cluster and analyze the potential sources of NOEEs and OEEs.

L315. Jones et al. 1999 is absent in the reference list.

Reply: The section 4.2 has changed a lot. The error has been changed.

L317-318. A very probable reason is that the DA station is higher and therefore closer to the tropopause.

Reply: The section 4.2 has changed a lot. The error has been changed.

L319. Do you mean the stratospheric polar vortex? Why do you mention it here? How is it related to ground level ozone?

Reply: The section 4.2 has changed a lot. The error has been changed.

L354-355. This explanation is unclear.

Reply: The fourth section has changed a lot. The original error no longer exists.

L359-369. This analysis is vague due to many reasons, see below.

Reply: The fourth section has changed a lot. The original error no longer exists.

L362-364. Bad language.

Reply: The fourth section has changed a lot. The original error no longer exists.

L263. September is not presented in Fig. 7.

Reply: The fourth section has changed a lot. The original error no longer exists.

L363-364. On what basis do you conclude about “extensive turbulence". The only source of turbulence in polar night is dynamical instability. But according to your data mentioned on page 15 the wind velocity was small during OEE events.

Reply: The fourth section has changed a lot. The original error no longer exists.

L363-365. I do not agree with this conclusion. Analysis of Figs 7a and c shows that there is no good correspondence between ozone maxima at Fig. 7a and subsidence of potential vorticity in Fig. 7c.

Reply: The fourth section has changed a lot. The original error no longer exists.

L365-366. The 50-200 hPa layer is not presented in Fig. 7.

Reply: The fourth section has changed a lot. The original error no longer exists.

L367. On what basis do you conclude that turbulence near the tropopause affects directly ozone? Do you really believe that there is intensive turbulence near the tropopause which is defined as a most statically stable layer?

Reply: The fourth section has changed a lot. The original error no longer exists.

L374. Which two events? The corresponding number is absent in the table.

Reply: The fourth section has changed a lot. The original error no longer exists.

L376-377. It is obvious, by definition of OEE, that increase during OEE is larger.

Reply: The fourth section has changed a lot. The original error no longer exists.

L380. What is PBLs? And what do lower PBLs mean?

Section4.3.3. Do not confuse vorticity with vortex.

Reply: The fourth section has changed a lot. The original error no longer exists.

L402-403. Negative value cannot be larger than positive value.

Technical corrections

Reply: The fourth section has changed a lot. The original error no longer exists.

L16. from -> at

Reply: The error has been changed.

L16. Specify that the Zhongshan Station is coastal.

Reply: Zhongshan Station ->Zhongshan Station (Southeast coast of Prydz Bay)

L28. “account for" is probably wrong word.

Reply: account for-> expound

L100. monitored -> measured

Reply: The error has been changed.

L104. spatial temporal -> spatial and temporal

Reply: The error has been changed.

L115. Give here the full name of the station.

Reply: The error has been changed.

L118. transported -> transferred

Reply: The error has been changed.

L123. related coefficients -> appropriate correlation coefficients

Reply: The error has been changed.

L178. experienced -> is characterized by

Reply: The error has been changed.

L192. stable -> less changeable

Reply: The error has been changed.

L193. variable -> more changeable

Reply: The error has been changed.

L309. pressure altitude?

Reply: The error has been changed. Pressure altitude corrected to pressure height.

L332. What is SI?

Reply: The words are wrong here. SI corrected to STT.

L336. stratosphere intrusion -> stratospheric intrusion

Reply: The error has been changed

L337. stratospheric-affected -> stratosphere-affected

Reply: The error has been changed

L340. define -> determine

Reply: The error has been changed.

L357. transmission -> transport

Reply: The error has been changed.