Response to Referee #2

The authors sincerely thank Reviewer #2 for the valuable comments and the very helpful considerations, which greatly contribute to an improvement of our paper.

In the following, we address the particular issues raised by Reviewer #2:

Major:

Q2.1: Until the description of modelling section there is this confusion that
1. What is the mechanism that drives ODEs?
2. What are the chemistry associated with this?
3. Are you talking about ODEs or TOC decrease?
Therefore, a careful rephrasing is needed in some places to clarify these doubts.

A2.1: Thanks a lot for the comment. To clarify the confusion of Reviewer #2, we made the following modifications to the original manuscript.

1. The mechanism driving ODEs is that in the presence of sunlight, the bromide ions stored in substrates such as ice-/snow-covered surfaces are activated by HOBr molecules in the atmosphere and then released into the atmosphere. The elevated bromine in the atmosphere then depletes the surface ozone in polar regions. We added more details in the Section "Introduction" to explain the mechanism driving ODEs. Please see **lines 44-45** and **lines 54-56** of the revised manuscript.
2. We actually included the description of the chemistry associated with ODEs in the original manuscript. Please see **lines 43-54**. In the revised manuscript, we added more explanations so that the readers can understand the chemistry more easily.
3. We distinguish them (ODEs and TOC decrease) more clearly in the revised manuscript. Please see **lines 62, 65 and 81**. Thanks a lot for the advice.

Minor:

Q2.2: L 1-2: I do not understand this. TOC influences ODES or the other way around?

A2.2: What we meant to express is that TOC influences ODEs. We have refined this sentence in the abstract. Thanks.

Q2.3: L 2: Usually "data" means measurements or observations (not model, reanalyses, etc.)

A2.3: Yes, the sentence here is not clear enough. We rephrased the related sentence. Please see **lines 2-4** in the revised manuscript.

Q2.4: L 7: seems? So how it happens? Just changes in TOCs?

A2.4: We used the word "seems" here because the conclusion is not definite according to the existing data. We have rephrased this sentence in the revised manuscript. Moreover, TOC is only one of the factors that can influence ODEs. To clarify the confusion of the reviewer, we modified the expression here; please see **lines 7-10** in the revised manuscript.

Q2.5: L 9-11: confusing .please state clearly

A2.5: Modified. Please see **lines 12-15**. Thanks for pointing it out.

Q2.6: L24: an extraordinary

A2.6: Corrected.

Q2.7: L 26: The measurements of TOC from these stations are described in detail by Kuttippurath et al., 2010. It's good to mention this here, so that the readers will get an idea about these station measurements.

Kuttippurath, J., Goutail, F., Pommereau, J.-P., Lefèvre, F., Roscoe, H. K., Pazmiño, A., Feng, W., Chipperfield, M. P., and Godin-Beekmann, S.: Estimation of Antarctic ozone loss from ground-based total column measurements, Atmos. Chem. Phys., 10, 6569–6581, https://doi.org/10.5194/acp-10-6569-2010, 2010.

A2.7: We added the reference and related discussions in the revised manuscript according to the suggestion of the reviewer. Please see **lines 35-36**.

Q2.8: L 33: it was first reported that

L 35; in the coastal

L 48: such as Temperature

L53: there is no observational evidence

L59: at the Arctic coastal stations

L 85: "figure out", use another word.

A2.8: All done. Thanks

Q2.9: L 119: It depends, not for all stations. Please rephrase

A2.9: The related contents have been modified in the revised manuscript. Currently, we use the TOC observations provided by two surface monitoring stations (i.e., the Halley station and the Faraday-Vernadsky station). Data from other stations nearby (Belgrano II and Marambio) were also adopted to ensure the representativeness of the TOC data from these two stations. We found that the correlation coefficient between the daily TOCs observed at the Halley station and the FAD station resides in a value range of 0.3-0.8. The difference between the observed TOCs at these two stations might be caused by atmospheric dynamics.

Please see **lines 126-140** in the revised manuscript. Thanks

Q2.10: L 121: What is out of scope, as long as Faraday station is in the study region?

A2.10: The content here has been modified. Please see **lines 137-140**. Thanks.

Q2.11: L 125: not really, they have longer periods of data. See Kumar et al., 2021, who have used same the data from those stations

Kumar P., J. Kuttippurath, P. von Gathen et al.: The increasing surface and tropospheric ozone in Antarctica and their possible drivers, Environmental Science and Technology, https://doi.org/10.1021/acs.est.0c08491, 2021

A2.11: We sincerely thank the reviewer for providing this information. According to this suggestion, we found more data of surface ozone from observation stations in the Antarctic, which can be used for a further investigation. Please see the modifications marked in red in **lines 144-148** of the revised manuscript. The reference recommended by the reviewer is also cited in the revised manuscript.

Q2.12: L 136: What kind of extreme events?

A2.12: We modified the expression here and added more explanations about the selection criterion used in this study. Please see **line 157** of the revised manuscript. Thanks.

Q2.13: L 154: reference for US standard atmosphere. How old is this reference table?

A2.13: We added the reference here. Please see **line 180**. Thanks.

Q2.14: L207: "Later, we present .."

A2.12: Corrected.

Q2.15: L277-278: "By doing that, we were able to figure out the impact on the occurrence of ODEs brought about only by the change in TOC using TUV and KINAL models."

TOC changes control the ODEs?

A2.15: TOC is only one of the factors affecting the occurrence of ODEs. The dominant mechanism that controls ODEs is actually the consumption by the massive bromine in the air due to the bromine explosion mechanism.

We modified the statement here in the revised manuscript. Please see **lines 334-336**.

Q2.16: L285-286: "Thus, from the model results, the decrease of TOC favors the occurrence of the tropospheric ODEs and the bromine release."

TOC releases bromine? You need to explain this in terms of chemistry not with the TOC changes. TOC change can happen with dynamics too

A2.16: The mechanism we derived in this study is that the decrease of TOC results in an increase of solar radiation especially the UV radiation reaching the ground surface. As a result, the rates of many photolysis reactions increase. As these photolysis reactions are important for the bromine release and the corresponding ozone depletion, the acceleration of these photolysis reactions thus lead to a speedup of the tropospheric ODEs. That is why we stated here that the decrease of TOC favors the occurrence of ODEs and the bromine release.

To clarify the confusion of the reviewer further, we added more explanations in the revised manuscript. Please see **lines 344-351**. Thanks.

Q2.17: Figure 2: There is an ant-correlation between TOC and ODEs, except for 2010, any reason?

In addition, you state that the ODEs accelerate when TOCs are smaller, not the other way around. However, the figure does not illustrate that (conclusions line 384-385). Please explain.

A2.17: According to the suggestion of Reviewer #1, we currently investigate the correlation between TOC and the occurrence of ODEs based on the **daily** values of TOC. As a result, the content mentioned here is largely modified in the revised manuscript. Please see the statements in **lines 253-268** of the revised manuscript.

The daily TOCs detected at Halley and Faraday-Vernadsky (FAD) as well as the surface ozone observed at Halley during the springtime of years 2007-2013 are shown in Fig. A1 of this rebuttal. The ODEs identified in this study are also marked in this figure.

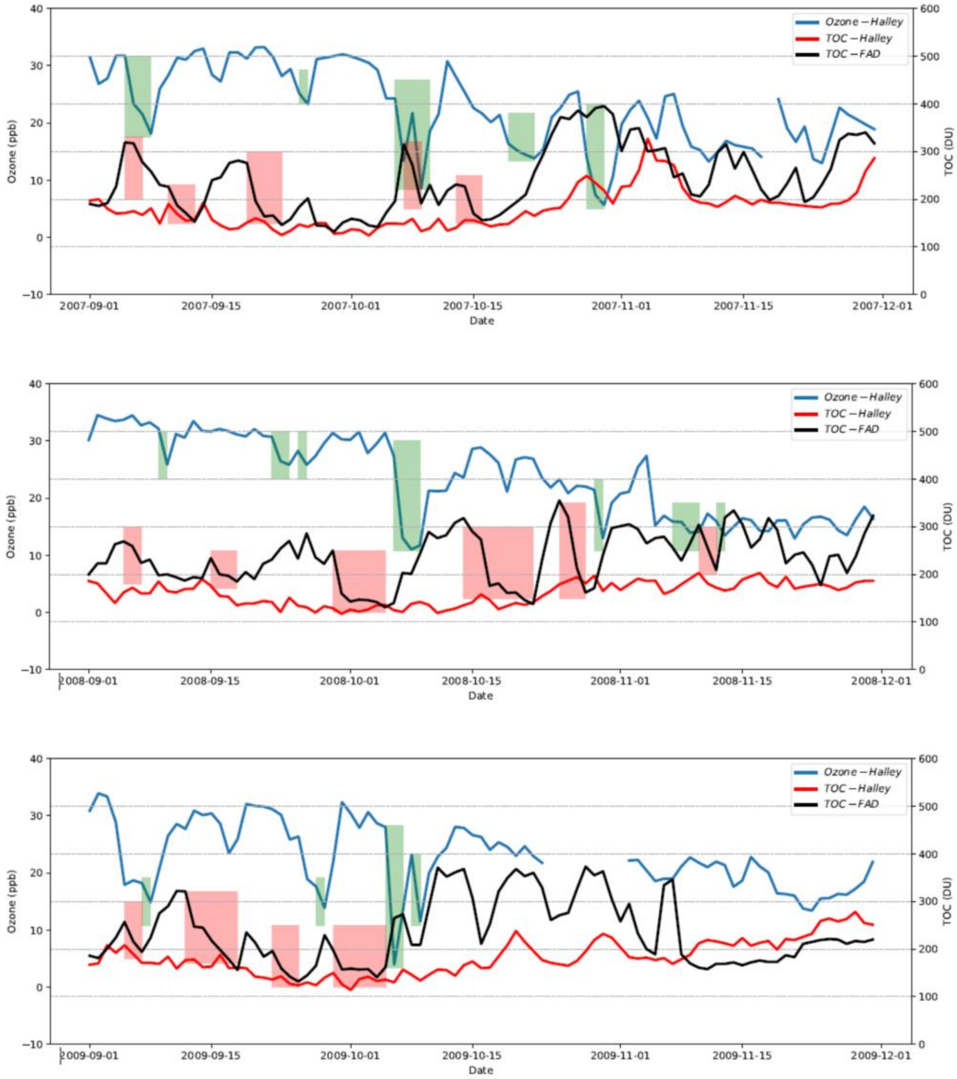

Fig. A1 (Continued…)

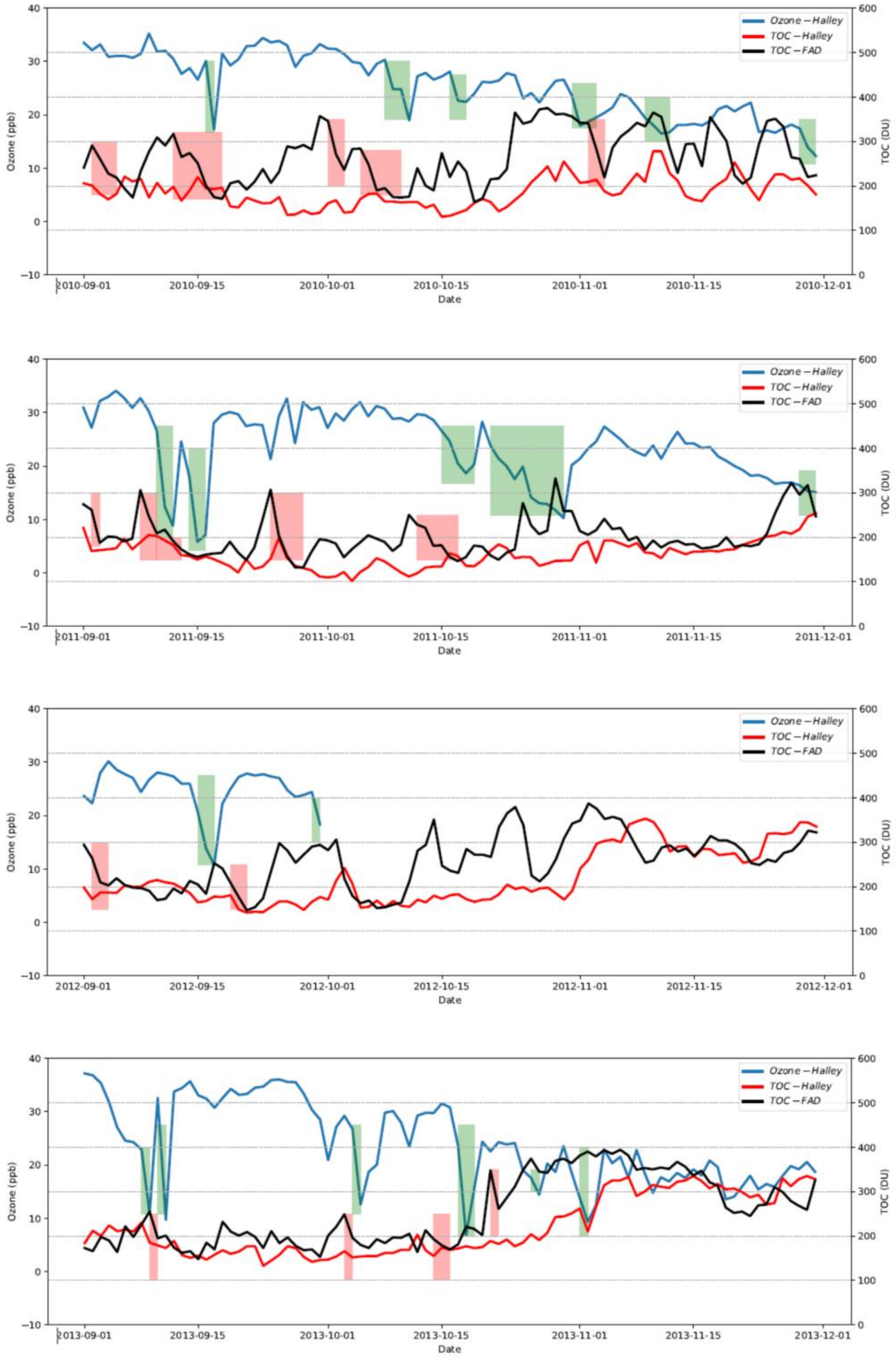

Fig. A1 Time series of TOCs belonging to the Halley station and the Faraday-Vernadsky (FAD) station as well as the surface ozone detected at Halley during the springtime of years 2007-2013 (the observational data of the surface ozone for October and November in the year 2012 are missing). The green-shaded areas in the figure indicate the periods identified as the occurrence of ODEs at Halley in the present study, and the red-shaded areas represent the significant decline in TOC at FAD, which might be associated with the occurrence of ODEs at Halley.

From Fig. A1, we found that the daily TOCs observed at these two stations (i.e., Halley and FAD) are different. We also calculated the correlation coefficients between TOCs observed at the Halley station and the FAD station, and the correlation coefficients mostly reside in a value range of 0.3-0.8. The difference between the observed TOCs at these two stations may be caused by atmospheric dynamics. Moreover, by comparing the surface ozone of Halley with the TOC detected at Halley (see Fig. A1), we did not find any obvious correlation between them, except that the ODEs occur more frequently in a relatively low TOC condition. However, from the comparison between the surface ozone of Halley and the TOC detected at the Faraday-Vernadsky (FAD) station, we found that the ODEs observed at Halley usually followed a decline of TOC detected at the FAD station (see the marks in Fig. A1). It suggests that the decrease of TOC over the area of FAD possibly favors the occurrence of ODEs at the Halley station. As the FAD station is located to the northwest of the Halley station and near the Weddell Sea (see Fig. A2 in this rebuttal for locations of the FAD station and the Weddell Sea), the TOC detected at this station is more capable of reflecting conditions of the Weddell Sea. Thus, we suggest the possible mechanism as that the decline in TOC over the area of the Weddell Sea favors the tropospheric ozone depletion in this region. Then the ozone-lacking air was transported from the sea to the Halley station, leading to the detection of ODEs at this site. Thus, there exists a lag time between the TOC decline observed at the FAD station and the detection of ODE at the Halley station, and the length of the lag time depends on the weather conditions during that period. In previous studies, the source of ODEs observed in Halley has also been discussed by by Jones et al. (2006), who found that air masses causing rapid ODEs of Halley originated in the southern Weddell Sea. Our findings are consistent with the conclusions of Jones et al. (2006).

Fig. A2 Locations of the Faraday-Vernadsky (FAD) station and the Weddell Sea in the Antarctic.

Due to the replacement of the monthly averaged TOC by the daily TOC observed at different stations in this study, different simulations and sensitivity tests were performed in the revised manuscript. We also modified the contents about the description of the observational data and the discovery of the relationship between TOCs and the surface ozone of Halley in the revised manuscript. Please see **Section 2.1.1** and **Section 3.1** for the refined results and related discussions.

References:

Jones, A. E., Anderson, P. S.,Wolff, E.W., Turner, J., Rankin, A. M., and Colwell, S. R.: A role for newly forming sea ice in springtime polar tropospheric ozone loss? Observational evidence from Halley station, Antarctica, Journal of Geophysical Research: Atmospheres, 111, 2006.