Response to Reviewer #1

We would like to thank Reviewer #1 for the careful and valuable comments, which greatly improve our paper. In the following, we address the comments of Reviewer #1 point by point.

Major:

Q1.1: What are ODEs, is it driven by chemical or physical (meteorology for instance) process? You mix of both these in this study in explaining this. For example, In Abstract it sounds that it is a meteorological phenomenon, but it's a chemical process, as detailed in Introduction. Please state what ODE is and how you define it in your study.

A1.1: The Ozone Depletion Events (ODEs) are phenomena in which ozone declines from the background level to a near-zero value in polar spring, which has been described in lines 26-27 in the revised manuscript. In general, the ODEs are initiated by the halogen chemistry including gaseous and heterogeneous reactions. Moreover, under stable stratification conditions in the Arctic region, the low-ozone situation can last for a few days or even longer, resulting in a long-distance transport of ozone-poor air masses. Essential meteorological conditions for the occurrence of ODEs include the sunlight, bromine-containing surface, and strong conversion on the top of the boundary layer, which has been proposed by Lehrer et al. (2004) and presented in lines 57-61 in the revised manuscript. Thus, the ODEs observed at a fixed station such as Barrow can be determined by the halogen chemistry, meteorological conditions, or both of them.

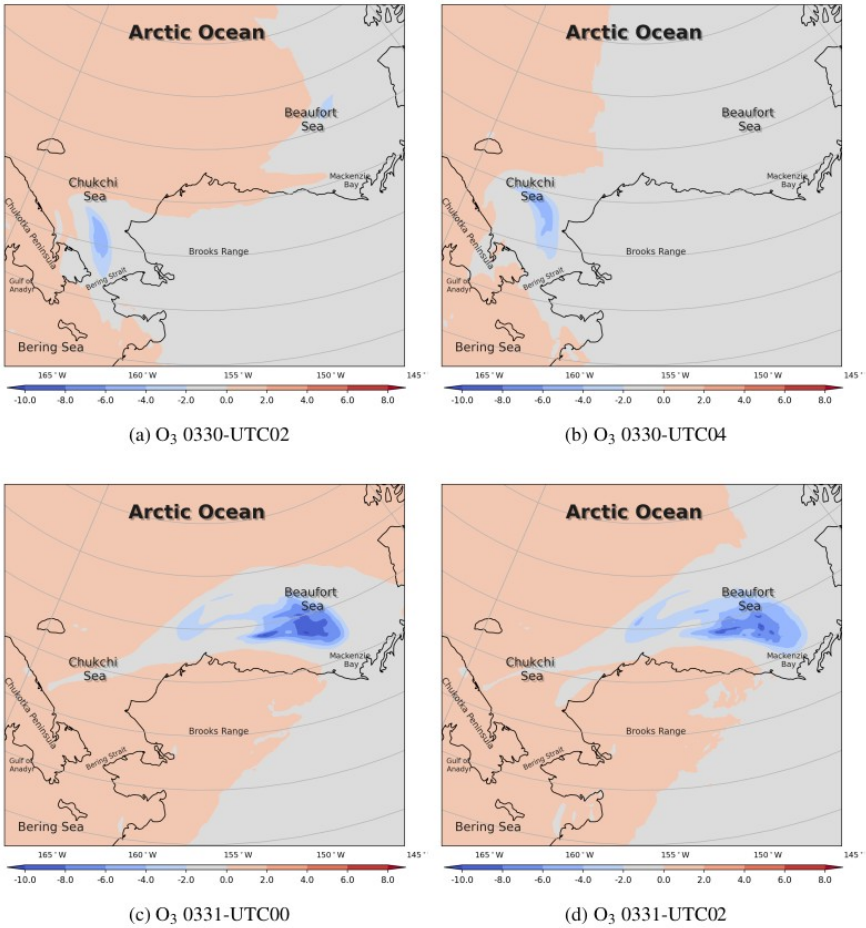
In our study, major factors for the occurrence of ODEs in different places (at Barrow and over the sea) are also different. For example, during ODE1, due to the formation of the cyclone, a large amount of sea-salt aerosols were released into the atmosphere. The liberation of sea-salt aerosols was able to emit bromine which can deplete ozone over the sea. Thus, in addition to the meteorology, the ozone depletion over the sea was also found to be largely contributed by the halogen chemistry. In contrast, at Barrow, the detection of ODE1 was mainly caused by a horizontal transport of low-ozone air from the Beaufort Sea to Barrow. Thus, ODE1 observed at Barrow was found to be determined by the meteorological conditions, especially the horizontal transport. To clarify the confusion of the

reviewer, we differentiate ODEs in different places more clearly in the revised manuscript. Please see lines 487-492.

Lehrer, E., Hönninger, G., and Platt, U.: A one dimensional model study of the mechanism of halogen liberation and vertical transport in the polar troposphere, Atmospheric Chemistry and Physics, 4, 2427–2440, https://doi.org/10.5194/acp-4-2427-2004, 2004.

Q1.2: In results, you said ODE1 is chemistry-driven and ODE2 is meteorology driven and just opposite is said in Conclusions in lines 359-364. Please clarify.

A1.2: This question is similar to **Q1.1**. In our study, we found ODE1 over the sea largely contributed by the elevated bromine due to the release of sea-salt aerosols, and the ozone-depleted air was then horizontally transported from the Beaufort Sea to Barrow, leading to the detection of ODE1 at Barrow. Thus, the ozone depletion at Barrow was found to be mainly driven by the horizontal transport, but the ozone depletion at the Beaufort Sea was largely affected by the bromine chemistry, as shown in Fig. 1 of this rebuttal. In contrast, ODE2 observed at Barrow was found to be caused by a horizontal transport of low-ozone air from the Arctic Ocean, and the bromine chemistry only played a minor role during this ODE. In order to clarify the confusion of the reviewer, we stated the ODEs in different regions more clearly in the revised manuscript; Please see lines 487-495.



(a) O_3 0330-UTC02 (b) O_3 0330-UTC04

(c) O_3 0331-UTC00 (d) O_3 0331-UTC02

Fig. 1 The change of surface ozone (ppb) caused by local chemistry, i.e., bromine chemistry, from 30 March to 31 March, 2019. The positive value represents a chemical production of ozone, while the negative value represents a chemical consumption of ozone.

Q1.3: A discussion on the model and measurement differences should be provided with respect to the new chemical reaction added, and also with the previous studies without that particular chemical reaction in the model. Improvements should be discussed.

A1.3: Thanks for the suggestion. In the revised manuscript, we added a section (i.e., Section 3.4 Sensitivity tests) showing the uncertainties in simulation results caused by using different heterogeneous reaction rates and boundary conditions. Please see lines 383-423 in the revised manuscript. Reasons for the deviations between simulations and measurements were also discussed in lines 227-234 in the revised manuscript. In addition, according to the reviewer's suggestion, we added more discussions about future improvements of the model; Please see lines 500-507.

Q1.4: The model domain and terrain maps are already provided in Figure 1. Then why do you want to repeat this in Figures 5-8 and 10?

A1.4: Thanks for the comment. In the original manuscript, we added the domain and terrain maps to the end of figures to remind readers of the geographic names appearing in the discussions of the paper. However, to indicate it more clearly, in the revised manuscript, we removed the redundant maps but marked the names of these places in the figures. Please see Fig. 5-Fig. 14 in the revised manuscript.

Q1.5: Presentation quality has to be improved significantly.

A1.5: Thanks a lot for the suggestion. During the revision process, we tried our best to improve the quality of the manuscript and made many corrections. Please see the marks in red and blue all thorough the revised manuscript. Moreover, we will also use the typesetting and language copy-editing services provided by the ACP journal (Copernicus Publications) to improve the quality of the paper. Thanks again for the suggestion.

Minor:

Q1.6: L2: In this study

A1.6: Modified. Thanks.

Q1.7: L3 and elsewhere in the MS: 28 March, not March 28th

A1.7: All places are corrected. Thanks a lot for the comment.

Q1.8: L4-5 rephrase "analysed thoroughly through process analysis" and state what process is analysed

A1.8: Processes investigated in this study include horizontal transport, vertical transport, dry deposition and the overall chemical process. We added the details of these processes in the revised manuscript. Please see line 6.

Q1.9: L: "ozone lacking air" means no ozone in that air. It is not correct, there will be always some

ozone.

A1.9: We changed it to “low-ozone air” throughout the revised manuscript. Thanks for the suggestion.

Q1.10: L9: what is local chemistry here? Bromine?

A1.10: The words “local chemistry” here means “the overall chemical contribution to the ozone change, which is mainly associated with the bromine chemistry”. Please see line 13 in the revised manuscript.

Q1.11: L10-11: What is this ODE? Chemistry driven or Transport driven? Is it because of ozone lacking air transported from some other region? The entire abstract sounds that the ODEs are meteorological events, Is that so?

A1.11: As mentioned in our previous answers **A1.1** and **A1.2**, ODEs are initiated by the halogen chemistry including gaseous and heterogeneous reactions. But under favorable meteorological conditions, the ozone-poor air can travel a long distance and the low-ozone situation would last for a longer time.

In our study, ODE1, which the reviewer doubted, was found to be initiated by the halogen chemistry in places near the Chukchi Sea. The bromine-rich air was carried to the Beaufort Sea and the halogen chemistry played a role in consuming ozone along this pathway. Thus, the ozone loss over the Beaufort Sea was largely attributed by the halogen chemistry. In contrast, the detection of ODE1 at Barrow was mainly caused by a horizontal transport of low-ozone air masses from the Beaufort Sea to Barrow.

Q1.12: L19: an ozone depletion event or you mean “the ozone depletion event”?

A1.12: It should be “ozone depletion events”. We have rephrased this sentence. Thanks.

Q1.13: L20-27; says ODE is chemical

A1.13: This reaction cycle denotes the chemical mechanism which can lead to the decline of ozone with the involvement of bromine. However, the occurrence of ODEs also needs certain

meteorological conditions which are favorable for the bromine accumulation and the ozone entrainment inhibition.

Q1.14: L28: catalyst for what?

A1.14: Catalyst for the ozone depletion. We have rephrased this sentence to “In this reaction cycle, the total amount of bromine stays constant, which means the bromine plays as a catalyst for the ozone depletion”. Thanks.

Q1.15: L57: Zeng et al., year is missing

A1.15: Added.

Q1.16: L64, year missing with citation

A1.16: Added.

Q1.17: L65: Marelle et al., year?

A1.17: Added.

Q1.18: L68: Both studies

A1.18: Changed.

Q1.19: Figure 1 caption; write the name of that study region

A1.19: Added. Thanks.

Q1.20: L95: 9 km

A1.20: Modified.

Q1.21: L101; particulate matter or particulates or specify them

A1.21: Corrected.

Q1.22: L105: Is this equation different from other CTMs in calculating the chemical tendencies”?

A1.22: No. This equation is similar to that in other CTMs. But we want to show this equation in our paper because it covers most of the processes elucidated by the process analysis method. We added more explanations for this equation in lines 118-119 in the revised manuscript.

Q1.23: L110: "incorporated or implemented" not instrumented

A1.23: We changed it to "incorporated". Thanks for the suggestion.

Q1.24: L111: A complete list of

A1.24: Corrected.

Q1.25: L126: "and if it is at a coastal area.."

A1.25: Done. Thanks.

Q1.26: Figure 2: I do not see any connection to the local meteorology and ODEs here. For instance, ODEs are found in both low and high temperatures/pressures/wind speeds. So what does this figure communicate?

A1.26: This figure shows the temporal evolution of meteorological parameters and chemical species at Barrow in a typical month with a high occurrence frequency of ODEs. Yes, as the reviewer pointed out, the connection between the local meteorology and the occurrence of ODEs is not obvious during the investigated time period. However, the temporal change of meteorological parameters reflect the meteorological conditions during ODEs and the meteorological conditions may significantly affect the occurrence of ODEs. Furthermore, for a successful simulation of ODEs, the model needs to capture the change of meteorology accurately. Thus, measurements of meteorological parameters shown in Fig. 2 were used to validate the simulations. Explanations for showing Fig. 2 in our manuscript are given in lines 172-174.

Q1.27: L143: why don't you just write "Measurements" instead of observational data

A1.27: We replace the words "observational data" with "measurements" in many places. Thanks a lot for the suggestion.

Q1.28: L151, 225, 320: In the “Supplementary Information”, not in the “supplements”

A1.28: Corrected throughout the manuscript. Thanks.

Q1.29: L166: of the meteorology

A1.29: Modified.

Q1.30: L172-175: How can you say these are “accurate”?

A1.30: The correlation coefficient R is a widely used statistical parameter. When the value of R is larger than 0.7, it indicates a very strong positive correlation. In this study, the correlation coefficients R of pressure, temperature, horizontal components of the wind speed (U and V) are 0.991, 0.92, 0.881, 0.897, which are all very close to 1.0, indicating a high agreement between observations and simulations. The RMSEs of pressure, temperature, U and V also denote small deviations between observations and simulations. We added more explanations in the revised manuscript. Please see line 183 and lines 208-210. Thanks.

Q1.31: Figure 3: write the parameter names, not “meteorological field”

A1.31: We wrote the names of meteorological parameters instead of “meteorological field”. Please see the caption of Fig. 3 in the revised manuscript. Thanks for the suggestion.

Q1.32: Figure 4: Why the simulated ozone is about 13 ppb higher than that of the measurements? This suggests that the model has a problem in simulating the ODE. Also there is a 3-4 days lag in Bromine explosion and the associated decline in ozone or ODE

A1.32: The reviewer is correct saying that there still exist a fraction of mismatches between simulations and observations. For ODE2 on 2 April, the model overestimated the surface ozone by approximately 10 ppb. After performing many sensitivity tests (shown in Sect. 3.4 in the revised manuscript), we found this ODE at Barrow to be greatly affected by a transport of low-ozone air from the Arctic Ocean which is located to the north of Barrow. As a result, the simulation of this ODE is heavily influenced by the boundary conditions implemented at the northern boundary of the model. Although we have modified the boundary conditions based on observations (Bottenheim and Chan, 2006), which has been described in Sect. 2.1.2, the simulation results still show some

deviations from the observations, indicating that a future improvement of the implemented boundary condition is still needed. We added more discussions about the deviations between simulations and observations in the revised manuscript, please see lines 226-234. Also, future improvements of the model were extended in the revised manuscript, please see lines 500-507.

With respect to the 3–4 day lag, we think the reviewer might mean the ozone decline on 29 March and the bromine elevation between 30-31 March. However, these two events were not corresponded. The bromine elevation between 30-31 March was actually associated with the partial ODE on 31 March. In contrast, the ozone decline on 29 March was mostly determined by the boundary conditions of the model, so that we are not investigating it deeper in this study.

Bottenheim, J.W. and Chan, E.: A trajectory study into the origin of spring time Arctic boundary layer ozone depletion, Journal of Geophysical Research: Atmospheres, 111, https://doi.org/10.1029/2006JD007055, 2006.

Q1.33: L189: 40%

A1.33: Corrected.

Q1.34: L236: as shown in Fig. 7c

A1.34: Corrected.

Q1.35: L239-240: What do you mean by “twisted” here?

A1.35: The word “twisted” is inappropriate, and we changed it to “deformed” in the revised paper.

Q1.36: L241-242: “At UTC 06 on March 30th, the sunset occurred.” Please rephrase this

A1.36: We rephrased this sentence to “At 06 UTC on 30 March, the sun was setting”. Please see line 304 in the revised manuscript. Thanks.

Q1.37: L253: retarded?

A1.37: We changed the word to “delayed”. Thanks for pointing out this inappropriate word.

Q1.38: L266: “These bromine atoms then consumed the surface ozone, forming BrO.” Please rephrase this. A similar sentence is also there in lines 314-315.

A1.38: We rephrased these sentences. Please see lines 331 and 334 in the revised manuscript.

Q1.39: L269: “and the amount of HOBr is also increased …”

A1.39: Rephrased. Thanks.

Q1.40: L271-272: “Then, sun rose again. Br2 photolyzed rapidly and BrO was formed again. Combine these sentences

A1.40: We combined these two sentences together. Please see lines 337-338 in the revised manuscript. Thanks.

Q1.41: L275: delete “occurring”

A1.40: Done.

Q1.42: L361-363: In results section, L321-322, you said it was not a chemical process. It’s contradictory.

A1.42: This question is similar to the questions **A1.1** and **A1.2**. The reviewer might be again confused by our inappropriate descriptions in the original manuscript. For the two ODEs investigated in this study, ODE1 over the sea was largely contributed by the bromine activation, while ODE2 was mainly caused by a horizontal transport of the low-ozone air to the study region. As a result, the chemistry contributed at most 10 ppb to the ozone loss over the Beaufort Sea during ODE1, but the chemical contribution to the ozone loss during ODE2 was small. With respect to Barrow, the ozone decline during these two ODEs were all mainly caused by the horizontal transport of low-ozone air to this observational station. Thus, the dominant factor for the occurrence of ODEs at Barrow was found to be the horizontal transport. In order to clarify the confusion of the reviewer, we rephrased these sentences in the revised manuscript. Please see lines 470-480.