The authors thank the reviewer 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 the reviewer:

R3.1: The model appears to be running without any diurnal pattern, i.e., with fixed photolysis rates, a situation that I do not think is representative of springtime conditions in the about 70-80 degree North regions where these ozone depletion events are typically investigated. At the very least, the authors need to discuss limitations of this approach, and justify the use of the model in this configuration. How are photolysis rates determined? How are the timescales for depletion altered by this assumption? Can any chemistry occurring in nighttime or low sun conditions play a role (e.g,, through formation of reservoir compounds) that is not captured in the model?

A3.1: In this model, the average photolysis reaction rates are adopted which have been used in the previous study (Lehrer et al., 2004). These photolysis reaction rates are evaluated by using a three-coefficient formula with albedo = 1.0:

$$J = J_0 \exp\{b[1-\sec(c\chi)]\}, \qquad (1)$$

where χ is the solar zenith angle. The coefficients J_0, b, c are determined from the data from Röth's ART model (Röth 1992, 2002) at $\chi = 0°, 60°$ and $90°$. The coefficient values for the species involved in photolysis reactions are listed in Tab. 1. This table and the photolysis reaction rate calculation will be added in the revised manuscript.

Lehrer et al. (2004) state that "A comparison of results with a model run under the simulation of diurnal and monthly SZA variations provided no substantial difference in time scale of ozone depletion and absolute concentration levels of relevant species". In order to clarify the effects caused

Table 1. Coefficients for the photolysis reaction rate, Eq. (1).

Species	J_0 [s^{-1}]	b	c
O_3	$6.85\ 10^{-5}$	3.510	0.820
Br_2	$1.07\ 10^{-1}$	0.734	0.900
BrO	$1.27\ 10^{-1}$	1.290	0.857
HOBr	$2.62\ 10^{-3}$	1.216	0.861
H_2O_2	$2.75\ 10^{-5}$	1.595	0.848
HNO_3	$1.39\ 10^{-6}$	2.094	0.848
NO_2	$2.62\ 10^{-2}$	1.068	0.871
$NO_3 \rightarrow NO_2$	$6.20\ 10^{-1}$	0.608	0.915
$NO_3 \rightarrow NO$	$7.03\ 10^{-2}$	0.583	0.917
$BrONO_2$	$3.11\ 10^{-3}$	1.270	0.859
$BrNO_2$	$1.11\ 10^{-3}$	1.479	0.851
BrCl	$3.41\ 10^{-2}$	0.871	0.887
Cl_2	$7.37\ 10^{-3}$	1.204	0.863
ClO	$1.08\ 10^{-4}$	3.876	0.816
HOCl	$7.47\ 10^{-4}$	1.396	0.855
$ClONO_2$	$1.29\ 10^{-4}$	1.286	0.861
OClO	$2.61\ 10^{-1}$	1.058	0.872

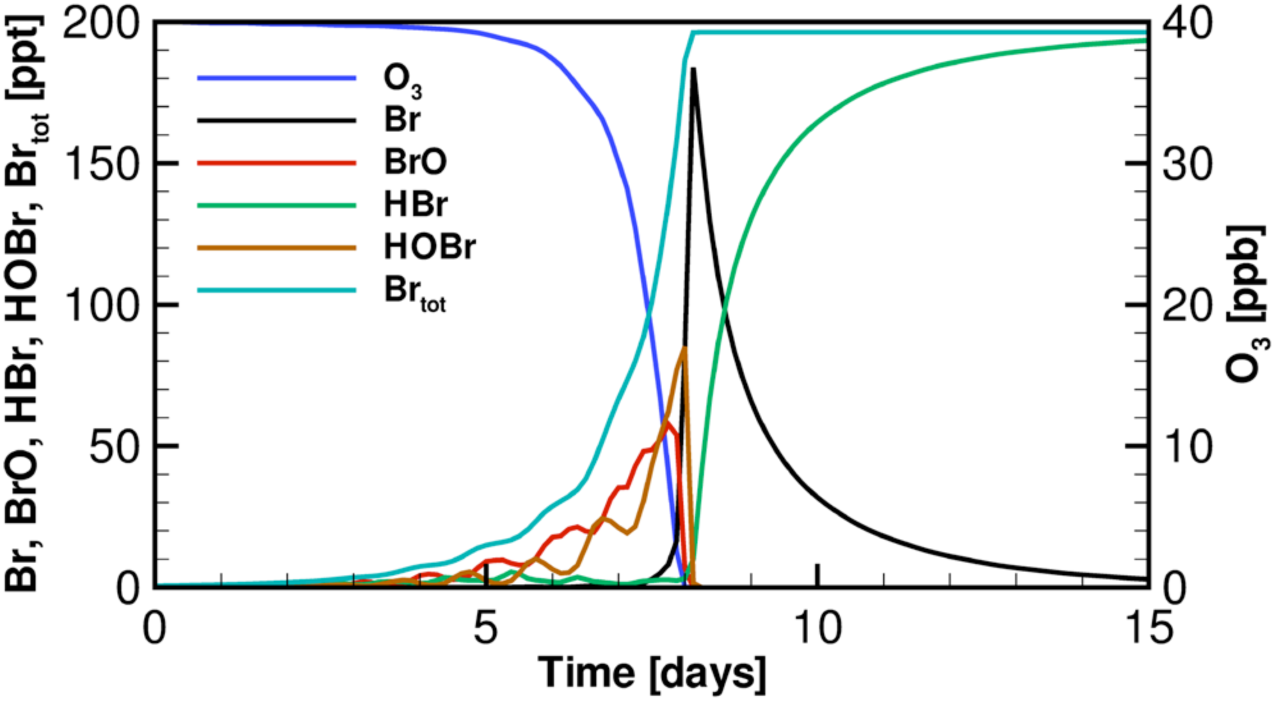

Fig. 1 Evolution of the chemical species concentrations with varying SZA in bromine only mechanism with 200 m BLH.

by the diurnal change, we conducted a simulation by using a varying SZA value. We use the initial χ_0 value of 85.5° for the date April 1 at 80° N. According to Lehrer's model, SZA at this location varies as

$$\chi = \chi_0 - 4.11E\text{-}6 \times \text{time} + 6.5 \cos(2\pi/86400 \times \text{time}). \qquad (2)$$

The first term in the RHS of Eq. (2) is the initial SZA value, and the second term represents the SZA change for different days in April. The third term denotes the daily change of SZA. This equation is used in an exploratory computation with varying SZA. Figure 1 shows the results, which can be compared to Fig. 1a of the paper. The variation of SZA has little effect on the temporal evolutions of the ozone and halogen species concentrations. The result shows a somewhat prolonged ozone depletion by about one day. This will be discussed in the revised manuscript.

The chemical reactions at nighttime or at low sun conditions may play some role as the reviewer suggests, this will be investigated in future studies. The present model aims to include the most important factors, and the model then can be refined with respect to effects that are considered of minor importance. We tried to make our model as complete as necessary and we could not think of any reservoir species, which could play a role.

R3.2: There are now measurements of HBr, HOBr and BrO from Barrow (Liao et al., JGR, 2012) – the authors should compare data from Figure 1 with this literature result.

A3.2: We will add the literature as the reviewer suggested. Unfortunately, the boundary layer height in the measurements and in the modeling study of Liao et al. (2012) is not provided. The mixing ratio of BrO is observed to lie between 20-30 ppt and HOBr around 20 ppt. From these values, we estimate that the boundary layer height in the measurements is about 1 km. In this situation, the comparison between their measurements and our computational results for the boundary layer height of 1 km is good, see Fig. 1c. This discussion will be added to the revised manuscript.

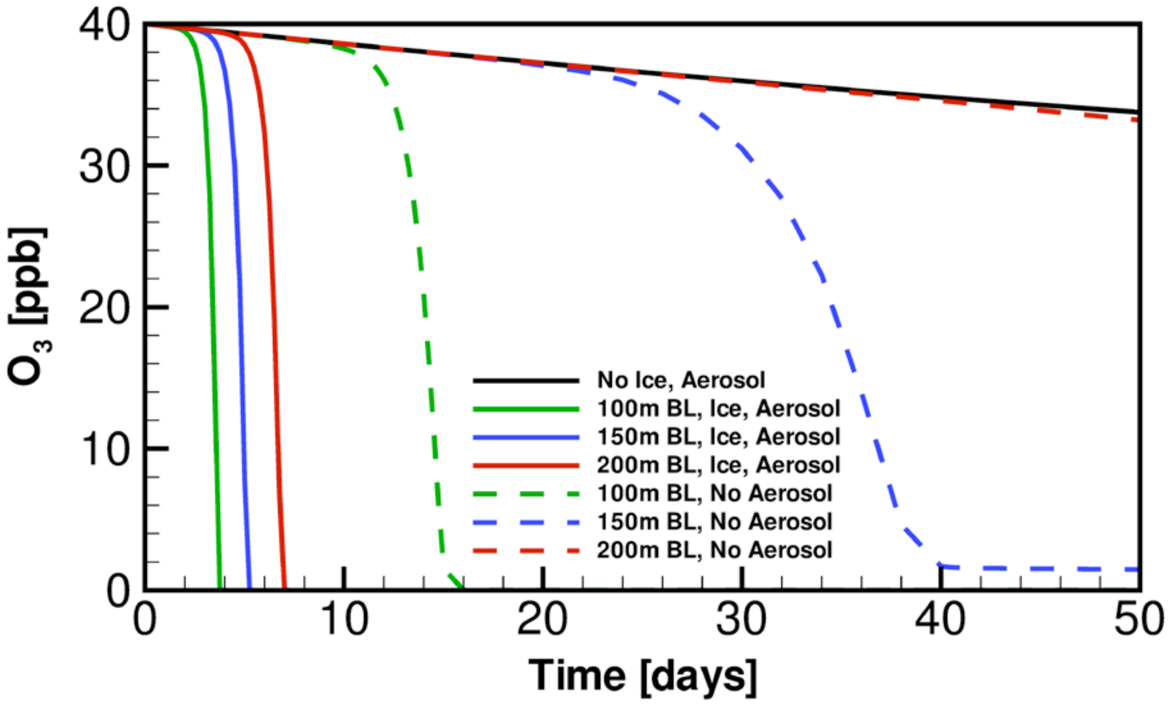

Fig. 2 Effect of heterogeneous reaction rate on the ozone depletion time.

R3.3: My understanding of this chemistry is that there is currently no consensus as to whether HOBr activation (R14, R15) occurs on aerosol or on the ice- or snow-covered ground. The results here seem to indicate that both are relevant – some further discussion seems warranted, particularly as a function of BL height since those tests were conducted.

A3.3: Exploratory simulations turning off the activation processes on aerosol or at the ice surface at different boundary layer heights were conducted to investigate the reviewer's question. The results of the simulations are presented in Fig. 2 for three different values of BLH. It is seen that both the ice surface and the aerosol play a significant role since both of them lead to strongly enhanced ozone depletion. The existence of ice surface is the major factor causing the ozone depletion, whereas the aerosol activation leads to an acceleration of the ozone depletion. This is because that the total halogen concentration is determined by the activation at the ice/snow surface, and the activation process on the aerosol surface helps to re-activate the halogen concentrations stored in HBr, thus, accelerating the ozone depletion event. It is also found that for a BLH of 200 m or higher, the ozone depletion event will not happen if either aerosol or ice surface activation is turned off. Thus, in the present simulations, both activation processes are needed. This result and the related discussion will be added in the revised manuscript.

R3.4: This is somewhat a case of semantics, but reaction (R5) and (R7) are referred to as a cycle, as are (R1) and (R3). However, these reaction pairs are really 'do-nothing' cycles, and it is really the difference in the rate of the reactions in each pair that determines whether ozone is destroyed or created. This point comes out to a certain extent, where differences in (R5) and (R7) are pointed out, but I would guess that (R1) and (R3) are nearly identical in rate and one could leave them out of the model entirely without any significant effect. Some further discussion or re-wording might be useful here. Also, I wonder (p. 24184, line 22-23) if it is HOBr formation from the BrO/HO2 reaction that has more effect in driving the difference between the rates of (R5) and (R7) than the formation of Br atoms in the BrO/BrO self-reaction.

A3.4: Reactions (R1) and (R3), (R5) and (R7) are reverse to each other. However, this does not mean that these reaction pairs are “do nothing” cycles so that they can be left out of the model. Let’s take (R1) $O_3 + h\nu \rightarrow O(^1D) + O_2$ and (R3) $O(^1D) + O_2 \rightarrow O_3$ as an example. The $O(^1D)$ produced in (R1) is not totally or directly converted back to O_3 in (R3), but other reaction steps in the complex reaction scheme (e.g. (R4) $O(^1D) + H_2O$) may also consume the reaction products. In fact, $O(^1D)$ produced in (R1) is the major source of the hydroxyl radical (OH). The OH concentration will affect the formation of HO_2 and HOBr, which is very important for the heterogeneous halogen release process and the ozone depletion rate. Therefore, removing these reactions from the chemical reaction scheme would lead to a very different ozone depletion behavior. This will be clarified in the revised manuscript.

We refer to reactions (R1) and (R3), (R5) and (R7) as “cycles”, since we want to know, in which reactions most of the ozone molecules are involved at different times. This does not mean that all the ozone is produced or consumed in these reaction pairs. For clarification, we will change the phrase 'reaction cycle' to 'reaction pair' in the revised manuscript as the reviewer suggested. It is true that the rate difference of these reaction pairs causes the decrease of the ozone.

We analyzed the contribution of various reactions and combinations of reactions to the ozone depletion at day 2, which is during the induction stage, cf. Fig. 3. For comparison, the same analysis is shown at

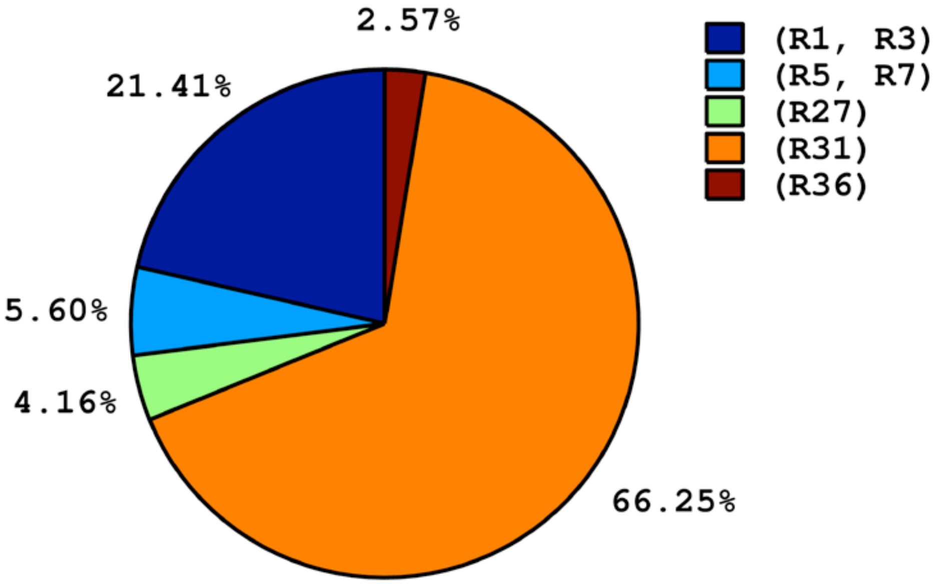

Fig. 3 Contribution of various reactions and combinations of reactions to ozone depletion at day 2.

(R1): $O_3 + h\nu \rightarrow O(^1D) + O_2$
(R3): $O(^1D)+O_2 \rightarrow O_3$
(R5): $Br + O_3 \rightarrow BrO + O_2$
(R7): $BrO + O_2 + h\nu \rightarrow Br + O_3$
(R27): $OH + O_3 \rightarrow HO_2 + O_2$
(R31): $HO_2 + O_3 \rightarrow OH + 2\ O_2$
(R36): $C_2H_4 + O3 \rightarrow HCHO + CO + H_2O$

day 6, which is within the ozone depletion stage, cf. Fig. 4. For an easier understanding of the figures, the reactions analyzed in these figures are listed below

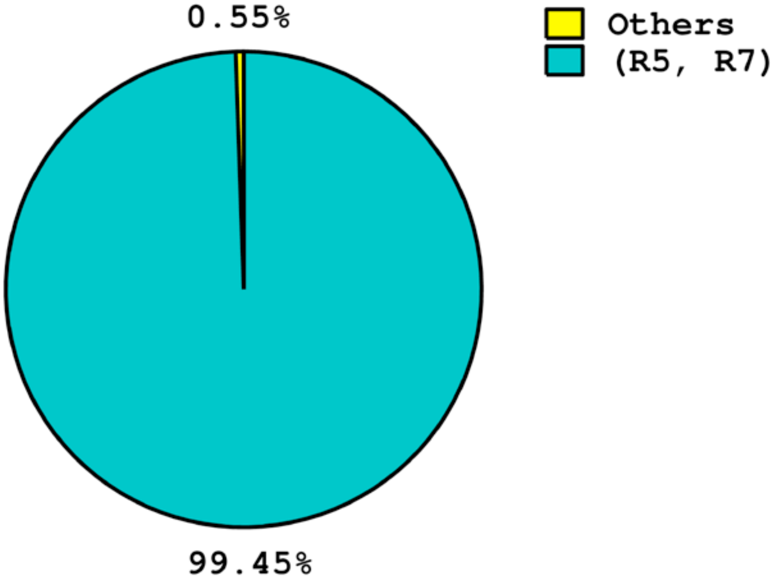

Fig. 4 Contribution of various reactions and combinations of reactions to ozone depletion at day 6.

(R5): $Br + O_3 \rightarrow BrO + O_2$
(R7): $BrO + O_2 + h\nu \rightarrow Br + O_3$

It is seen in Fig. 3 that during the induction stage, although most of the ozone molecules are involved in the reaction pair of (R1) and (R3) as discussed in the paper (p. 24184 L.24-26), the largest contribution to the ozone depletion attributes to (R31) HO_2+O_3->OH+2 O_2. On the contrary, during the depletion stage (cf. Fig.4), most of the ozone molecules are consumed by the rate difference between (R5) Br+O_3->BrO+O_2 and (R7) BrO+hγ+O_2 ->Br+O_3. Therefore, we continue to figure out the reason for the rate difference of these two reactions (R5) and (R7) during the ozone depletion stage.

For the difference between the rates of (R5) and (R7) during the depletion stage, we found that the increase of the Br radicals is the major reason. This increase of Br radicals is caused by the BrO/BrO self-reaction (R8). It is confirmed by the result displayed in Fig. 5 in which the contribution to the production of Br concentration at day 6 (during the depletion stage) is shown. Further discussion will be presented in the revised manuscript.

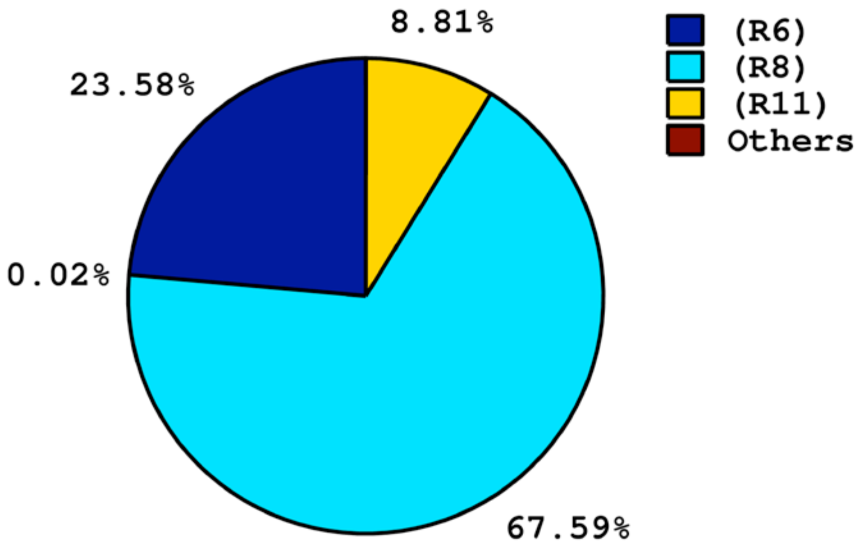

Fig. 5 Contribution of various reactions to Br formation at day 6.

(R6): $Br_2 + h\nu \rightarrow 2\ Br$
(R8): $BrO + BrO \rightarrow 2\ Br + O_2$
(R11): $HOBr + h\nu \rightarrow Br + OH$

R3.5: With respect to NO_X, cycle (N-I), is HO_2NO_2 in roughly steady-state with HO_2 and NO_2, such there is no net change in NO_X? Again, this is a do-nothing cycle, unless other chemistry is affecting things and the rates of (R62) and (R63) are not in balance.

A3.5: Similarly, an investigation of the major contributions to NO_X in the induction stage was done. The result at day 1 is shown in Fig. 6. It can be seen that in the induction stage, most of the NO_X molecules are converted to $BrONO_2$, thus leading to the increase of the $BrONO_2$ mixing ratio. This enhanced $BrONO_2$ mixing ratio will make the $BrONO_2$ involved reactions become more important in the depletion stage as discussed in the paper. This discussion will also be added in the revised manuscript.

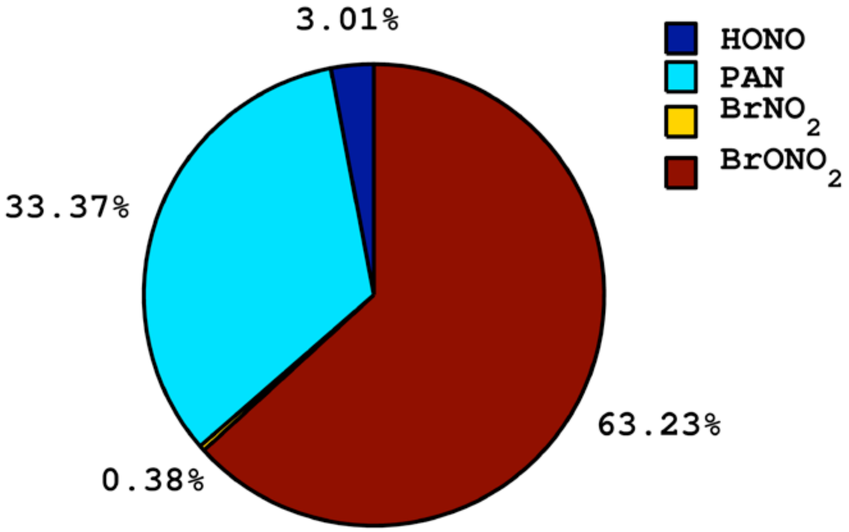

Fig. 6 Conversion of NO_X at day 1.

R3.6: On page 24189, it still seems that HNO_3 photolysis on surfaces will be very slow (life-time of a couple of days even if it is 100X faster than in the gas phase). Thus, it is not clear to me that the reaction cycle (I) is really representative of what is happening.

A3.6: From the observations it is found that the photolysis of HNO_3 at the surfaces will significantly increase than in the gas phase. However, this photolysis rate at the surfaces is difficult to determine. Thus, in the model, as described in p.24182, L.1-3 of the manuscript, we assumed that a similar constant value of uptake coefficient 0.06 can be used for this heterogeneous photolysis reaction $HNO_3 + hv \rightarrow OH + NO_2$ (R85). By using the values of the total aerosol surface area and gas phase molecular diffusivity in the model, the reaction rate constant is calculated as 3.30×10^{-4} s^{-1}. This is about 10,000 times faster than the rate constant for the gas phase. On page 24189, the work of Finlayson-Pitts (2009) is cited to prove that the increase of the HNO_3 photolysis reaction at the aerosol surface is possible, and the assumption used in the model is reasonable. However, a more precise parameterization of the photolysis rates of HNO_3 at the surfaces may be a topic for the future research. The corresponding paragraph will be rephrased in the revised manuscript.

R3.7: There is at least one previous discussion in the literature of the effect of NOx on ozone depletion events (e.g., Ridley et al., J. Atmos. Chem., 2003) – the results here should be compared and contrasted with that work. Also, the recent work of Stephens et al. (JGR, 2012) contains new information regarding Cl chemistry in polar regions, and this work could be discussed as well.

A3.7: We will add the contents for the comparison of our model results with the literature as the reviewer suggested. In fact, the temporal behavior of the NO_X mixing ratio in our simulations compares well with the study of Ridley et al. (2003), in which the NO_X mixing ratio is mostly between 10-20 ppt during the ozone depletion time. Our result for NO_X mechanism is also consistent with the measurements conducted by Beine et al., (2002a, b) at Alert in spring of the year 2000. In Beine et al., (2002a, b)'s measurements, during the ozone depletion stage, NO_X will decrease. As the ozone is almost totally consumed, the NO_X mixing ratio increases.

For the comparison of Cl chemistry, it is found that in our simulations, the gas phase Cl atom concentration has the value of about 0.6 10^4 molec. cm^{-3}. In the work of Stephens et al. (2012), it is found that the gas phase Cl atom concentration lies between 2.0 10^4 molec. cm^{-3} and 1.0 10^5 molec. cm^{-3}. The reason for this discrepancy might be the unknown chlorine source in their model as discussed in the manuscript (p. 24192, L11-14).

R3.8: I am confused by the concluding paragraph (p. 24194). If the heterogeneous reactions (i.e., HOBr reaction with HBr) are left out of the model during the induction phase, will it not be the case that one will never get to the depletion phase?

A3.8: The reviewer is right. Even though the heterogeneous reaction rate (e.g., $HOBr + H^+ + Br^-$) is quite low during the induction stage, it cannot be removed. We have checked that if the heterogeneous reaction is turned off during the induction stage, the time length of the induction stage will be extended and the depletion will be delayed. We had some misunderstanding about this before. This will be corrected in the revised manuscript.

R3.9: p. 24172, line 3 – might read better in the past tense – "...polar regions has been investigated..."

A3.9: The tense will be changed.

R3.10: p. 24173, line 19-20 – might read better as follows: "...since the 1990's, Box models..."

A3.10: The line will be changed as suggested.

R3.11: p. 24180, line 17 – 'considered' is spelled incorrectly

A3.11: This will be corrected.

R3.12: p. 24182, line 3-4 – might read better as follows: "...which photolyzes very slowly..."

A3.12: The line will be changed.

R3.13: There are a few places in the manuscript where what I would call obvious or repetitive statements are made. For example, the paragraph beginning on p.24185, line 26 could be condensed into a 'tighter' statement. Another place is with respect to PAN (on p.24188, line 11).

A3.13: The text at these two places will be rephrased.

R3.14: p. 24190, line 4 – It would be more accurate to state the following: "Peroxy radicals generated from volatile organic compounds (VOCs) could also replace HO2..."

A3.14: This line will be changed as suggested.

R3.15: One detail regarding the mechanism – it seems as though $O(^3P)$ and $O(^1D)$ atoms are being treated as one species. For example, NO_3 photolysis produced $O(^3P)$, but in the mechanism these atoms are 'allowed' to react with H2O to produce OH, which is not correct. (This probably is only a small effect, but something to perhaps change for future work).

A3.15: We thank the reviewer for pointing out this mistake. We have revised our mechanism and corrected the mistake. The corrections for the chemical reaction mechanism are:

(R1)	$O_3 + hv \rightarrow O(^1D) + O_2$
(R3)	$O(^1D) + O_2 \rightarrow O_3$
(R4)	$O(^1D) + H_2O$-> 2OH
(R69)	$NO_3 + hv \rightarrow NO_2 + O_3$
(R135)	$OClO + hv + O_2 \rightarrow ClO + O_3$.

Moreover, the product of the reaction OH + OH (+ M) (R54) is corrected to be H_2O_2.

The result using the revised chemical reaction mechanism for bromine only mechanism is shown in Fig. 7. It can be seen that these corrections make the computational results only slightly different from the ones shown in the manuscript.

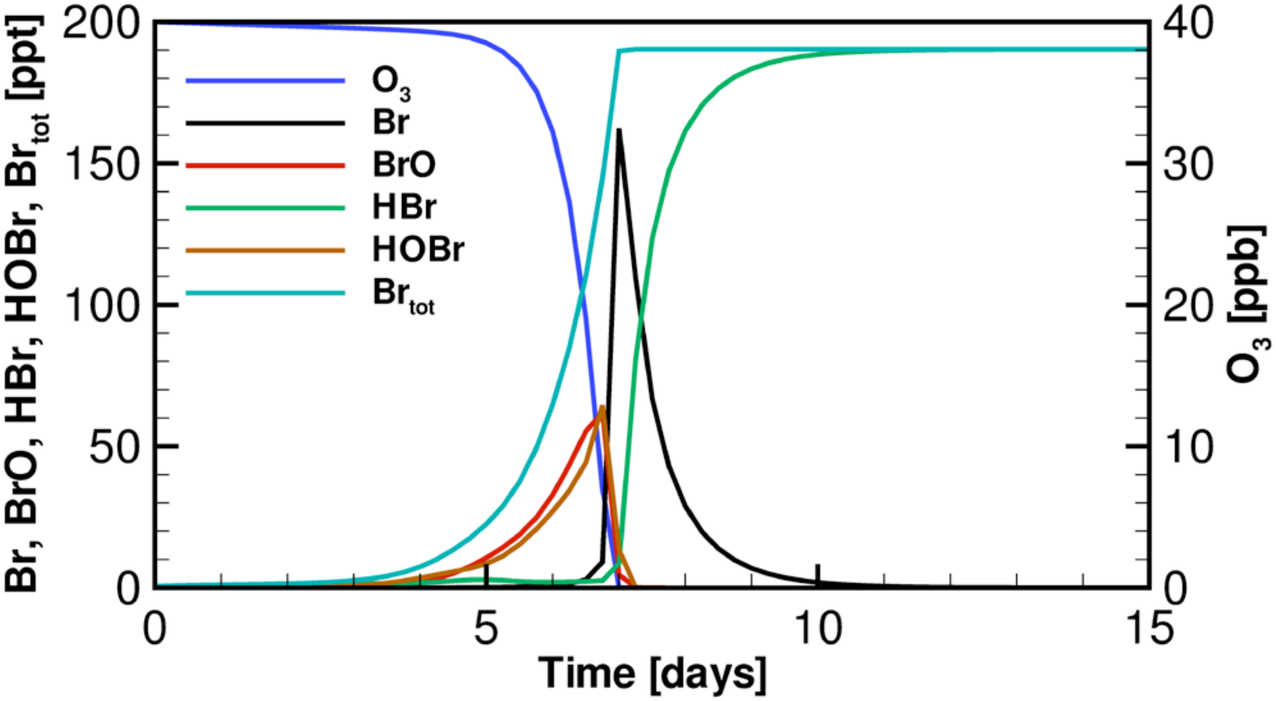

Fig. 7 Temporal variation of mixing ratio of ozone and bromine using the bromine only mechanism for the boundary layer height of 200 m.

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