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:

Major Comments:

R2.1: Introduction has been improved by dropping the redundant description of reaction cycles of potential significance, but it would read even better if the authors correct the following inaccuracy and incoherence in the context of cited studies: (1) all the papers cited at P24173, L9-10 only reported the measurements of XO radicals unlike what is indicated here by the authors, so the list of papers should be expanded in regard to halogen atoms, molecular halogens, etc.; (2) the work by Sander and Crutzen (1996) is not directly related to halogen activation in polar regions and should be dropped from citation at P24173, L21 and P24174, L2-3; (3) after all, the authors run the model at T = 258 K, which is above the eutectic of both NaCl-H2O and NaBr-H2O (e.g., Koop et al., 2000), making their argument in the paragraph of P24174, L1-13 rather irrelevant; (4) studies performed by the 1-D model MISTRA-SNOW (Thomas et al., 2011) are referred to as an example of coupled snow-gas phase chemistry "intensely studied" via modeling, right after a paragraph in which its methodology (the assumption of a liquid-like layer) is criticized; and (5) major difference between cited 3-D model studies lies in the types of halogen sources (wind-blown snow over sea ice – Yang et al., wind-blown frost flowers from refrozen leads – Zhao et al., snowpack over sea ice – Toyota et al., or using satellite BrO measurements not by asking what the source is – Zeng et al.) rather than the treatment of turbulence in the model.

A2.1: (1) The list of the references is expanded as requested. (2) Citation of Sander and Crutzen (1996) will be removed from both of these two places. (3) The paragraph will be rephrased in the revised paper. (4) We will remove the sentences in which the assumption of a liquid-like layer is criticized. Moreover, we will add the references (Abbatt et al., 2010; Oldridge and Abbatt, 2011), which support the important role of the liquid-like layer on the salty ice surface as the reviewer suggested. In addition, we will add a paragraph discussing the purpose of the present study and the reason for choosing the first order parameterization for the heterogeneous reaction as follows:
"The major focus of this study is the relative importance of each reaction and how the key parameters such as the boundary layer height affects the time scale of the ozone depletion event. Thus, a simplified treatment of the heterogeneous reaction is adopted in the model". See also the answer **A2.2**.

(5) The description for the development of the numerical models will be rewritten as follows:

"To date, a number of numerical studies have been conducted. Box models were used to capture the temporal evolution of the chemical species and investigate the recycling of halogen species on the surface of aerosol or ice covered by snow (Fan and Jacob, 1992; Tang and McConnell, 1996; Sander et al., 1997; Michalowski et al., 2000). More recently, box model studies (Sander et al., 2006; Morin et al., 2008; Sander and Morin, 2010) focus on the role of calcium carbonate precipitation in the triggering of bromine explosion and propose an application range of their conclusions. The coupling of HO_X, NO_X and halogen chemistry is also investigated (Chen et al., 2007; Bloss et al., 2010; Liao et al., 2011). In these box models, different techniques scenarios have been applied explicitly or implicitly to include the key heterogeneous reactions occurring at the aerosol surface or at the snow/ice surface.

Models with higher dimension are constructed. Lehrer et al. (2004) propose the primary source of the reactive halogens to be the sea ice surface by using a 1-D model. The coupled snow-gas phase chemistry is intensely studied by Thomas et al. (2011, 2012) using a 1-D snow-atmosphere model (MISTRA-SNOW) developed from the 1-D atmospheric boundary layer model MISTRA (von Glasow et al., 2002a,b; von Glasow and Crutzen, 2004; Piot and von Glasow, 2008, 2009). In the MISTRA-SNOW model simulation, special attention is paid to the interaction between bromine and nitrogen containing species over the snow-covered regions. The importance of the bromine re-activation on aerosol and snow is also addressed. Zeng et al. (2003, 2006) firstly implement a three-dimensional model to estimate the depletion of ozone using the BrO measurements derived from the satellite VCD (vertical column density). Zhao et al. (2008) propose that the aerosols from the frost-flowers might be the major source of reactive halogens using a global three-dimensional model. Yang et al. (2008, 2010) show that when a stormy blowing-snow condition occurs, bromine is mainly derived from the wind blown snow particles during the ozone depletion event. It is confirmed by the measurements conducted by Jones et al. (2009, 2010) in the Antarctic, concluding the weather conditions for which the ozone depletion event is more likely to occur. The snow pack is suggested as a potential source of halogen species (Toyota et al. 2011, 2013) using different models to analyze the air-snow pack interaction. In these models, the coupling of bromine chemistry and wind speed as well as air mass temperature is parameterized, addressing the connection between the bromine explosion, the ozone depletion, and the local atmospheric conditions."

R2.2: Given our poor understanding of physical chemistry of natural snow in regard to halogen activation, I do not feel that the authors are obliged to explain too much why they choose to represent halogen activation from the snow entirely by the ad-hoc choice of gamma values for HOBr and BrONO2. Logics used by the authors to justify their approach in the fourth paragraph of Introduction just do not convince me enough, especially because there are also experimental studies that indicate the roles of gaseous ozone uptake and nitrate photolysis (as OH source) for the release of gaseous bromine presumably from the liquid-like layer on the salty ice surface (e.g., Abbatt et al., 2010; Oldridge and Abbatt, 2011). I already see the value of the paper in the analysis of how key metrics such as Lmix and beta affect the timescale of ozone depletion. So it would sound more compelling if the authors stated that they took a minimalist approach for the present purpose.

A2.2: We like to thank the reviewer for the suggestion. After identification of a suitable chemical reaction mechanism, the goal of the present study is to investigate the effects of the meteorological properties such as boundary layer height and reactive surface ratio. The sensitivity analysis helps to show the relative importance of each reaction and the mentioned properties in the model. Based on the results obtained by the sensitivity of the chemical reaction steps, it is possible to reduce the size of the chemical reaction mechanism without losing important information. The skeletal mechanism may be used for multi-dimensional computations (L. Cao and E. Gutheil, 2013). We will add information about the purpose for the use of this simplified approach for the treatment of heterogeneous reactions. As the major focus of this study is the relative importance of each reaction and the effects of the weather conditions such as the boundary layer height, a simplified treatment of the heterogeneous reaction is adopted. The results show that even though the treatment is relatively simple, the major features of the ozone depletion and halogen release phenomenon can still be captured. In principle, it is possible to include the liquid reactions, porous snow pack and the mass exchange between the liquid phase and gas phase to the present model, which will be the focus of future work. The limitations of the model will be stated in the revised manuscript as follows:

"A photo-chemical box model is developed. The major focus of the present study is on the relative importance of each reaction and how the key metrics such as the boundary layer height affect the time scale of the ozone depletion event. Thus, a simplified treatment of the heterogeneous reaction is adopted. It is assumed that the saline snow/ice surface is the source of the halogen species. The collisions between HOBr molecules from the overlying atmosphere and the saline surface determine the liberation of Br_2 and BrCl from the surface back to the surroundings. The heterogeneous reactions occurring at the aerosol surfaces help to re-activate the inert halogen from the hydrogen halides".

R2.3: The parameterization described in Section 2.2.2 implies that the net source of halogens exist entirely at or very close to the bottom of the atmosphere. I will not criticize the authors for limiting their scope by this choice, but they should be cautious when they talk about the source of halogens in the model. It is not fully appropriate to indicate that this model accounts for sea-salt aerosols and/or wind-blown snow as a source of halogens (e.g., P24183, L2).

A2.3: We will rephrase the sentences stating that sea-salt aerosols are considered as a source of the halogen species. However, the heterogeneous process at the aerosol surface could re-activate the halogens stored in HBr, which may accelerate the ozone depletion event. To clarify this, the role for these activation processes at the ice/snow surface and aerosol surface is investigated by turning off the activation processes on the aerosol or at the ice surface at different boundary layer heights as shown in Fig. 1. It is found that the occurrence of the ozone depletion event is determined by the halogen concentration activated from the ice/snow surface. The re-activation process on the aerosol surface helps to shorten the induction time. It is also found that for a BLH of 200 m or higher, turning off either of these two activation processes, the ozone depletion event will not happen (at least not within 50 days), which means that both activation processes are needed for a 200 m boundary layer or higher.
It is possible to extend the parameterization of the aerosol by considering the initial halogen concentration in the suspended aerosols in the boundary layer so that the aerosols can be included as a source of the halogens in the model, but the typical concentration of halogen ions in the aerosols needs validation, and this will be done in future.

R2.4: It would be useful if the authors could differentiate between "induction stage" and "depletion stage" in a more objective manner. Currently, the depletion stage is loosely defined as "the depletion rate is in the order of 1 ppb h-1" (P24184, L1-2) and the authors seem to reply on the eye-ball examination of time-series plots for the assignment of the stages. I wonder if the authors could boldly define an exact threshold for the rate of ozone decrease in ppb per hour to differentiate between the

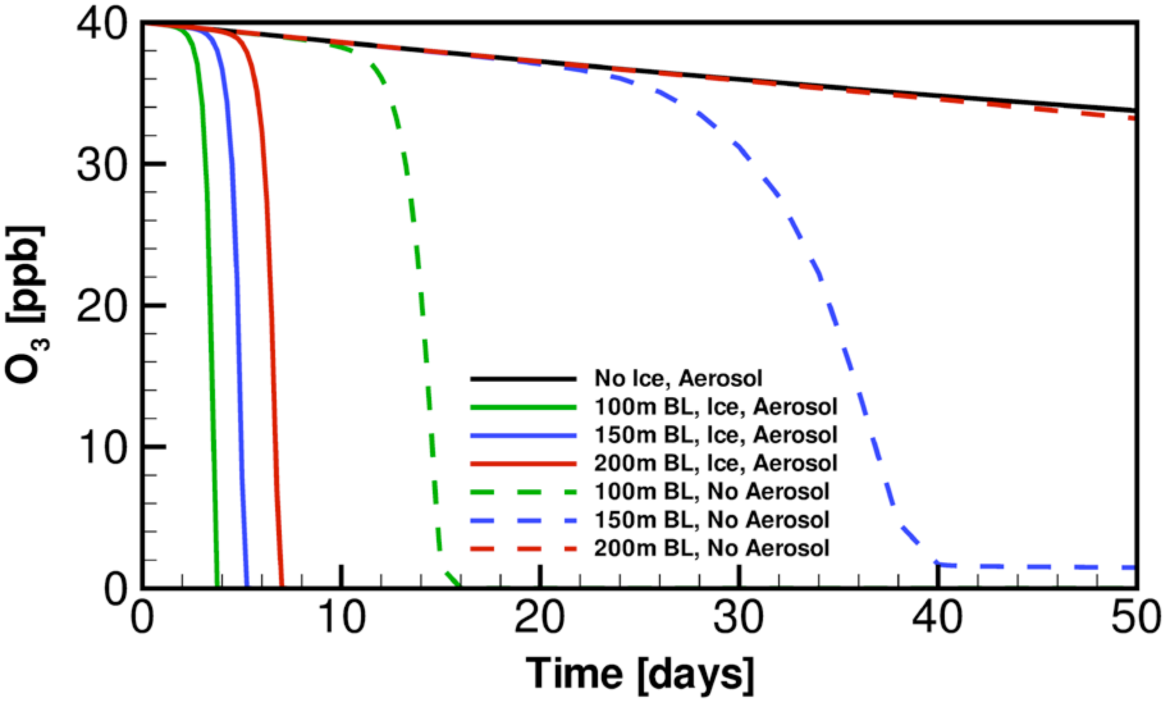

Fig. 1 Effect of heterogeneous reaction rate on the ozone depletion time.

"induction stage" and the "depletion stage". That way, the authors' discussion on the timescale of simulated ODEs would become more compelling, particularly for cases with shortened ozone depletion timescales by using high beta values. How long do the "induction" and "depletion" stages last respectively where the beta value approaches 100 and thereby the simulated ozone depletion is completed in about 2 days from the initiate state?

A2.4: We define the "depletion stage" as the time period in which the ozone depletion rate is larger than 0.1 ppb per hour. Moreover, we consider the end of the depletion stage as the time when the ozone mixing ratio is less than 4 ppb (i.e. 10 % of the initial mixing ratio). According to these definitions, the time for the "induction stage" and "depletion stage" is about 5.1 days and 1.75 days for 200 m boundary layer height, 16.8 days and 2.7 days for 500 m boundary layer height, 32 days and 4.25 days for 1000 m boundary layer height. This will be added in the revised paper. With these definitions, we improved the figure showing the value of beta on the time scale of the ozone depletion event for 200 m boundary layer height. The result is presented in Fig. 2. It is seen from Fig. 2 that as the value of β increases, the time needed for the induction stage significantly decreases, whereas the time for the depletion stage is approximately constant. When the beta value approaches 100, the induction stage is about 28 hours and the depletion stage 20 hours. This result will be added to the revised manuscript.

R2.5: I agree with Referee #3 in that the analysis of ozone budget (P24184, L10-28) is performed somewhat in a different way than we usually learn at the school of atmospheric chemistry. We usually group species that are coupled via fastest reaction cycles and look at the net gain or loss of the grouped species. In the present case, I would rather look at NO + HO2, CH3O2, etc. as gross ozone formation reactions (but NO is absent in this run and therefore the gross ozone formation would be marginal) and BrO + BrO, BrO + HO2, HO2 + O3, etc. as gross ozone loss reactions. If the authors want to stick to their current approach, I would like to see some more explanation by them as to why they choose to discuss the photochemical ozone budget in the way as currently done. Also, the inaccuracy of

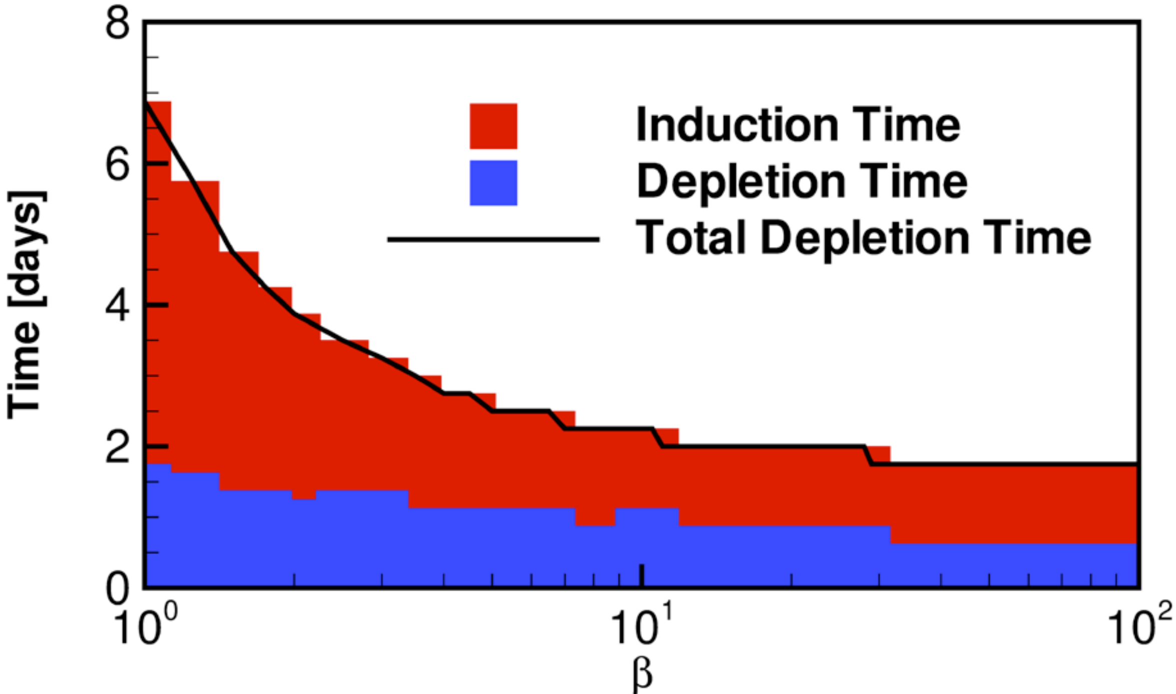

Fig. 2 Impact of the reactive surface ratio on the total ozone depletion time, the induction and the depletion time for a 200 m BLH.

representation for the species “O” in the model (see the major comment #7) makes this paragraph quite confusing. I see the same kind of species grouping problem in Section 3.2 where the authors discuss the NOx budget (P24187, L14-27).

A2.5: The change of the ozone mixing ratio is calculated as

$$d[O_3]/dt = -r_{R1} + r_{R3} - r_{R5} + r_{R7} - r_{R27} + r_{R30} - r_{R31} - r_{R36},$$

where r is the reaction rate of the following reactions:

(R1) $O_3 + h\nu \rightarrow O(^1D) + O2$

(R3) $O(^1D) + O_2 \rightarrow O_3$

(R5) $Br + O_3 \rightarrow BrO + O_2$

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

(R27) $OH + O_3 \rightarrow HO_2 + O_2$

(R30) $OH + OH + O_2 \rightarrow H_2O + O_3$

(R31) $HO_2 + O_3 \rightarrow OH + 2O_2$

(R36) $C_2H_4 + O_3 \rightarrow HCHO + CO + H_2O$.

We did not group species, since the importance of different chemical reaction steps is investigated. The reaction rates at different time periods are studied to see, in which reaction pairs most of the ozone is involved. This procedure enables the judgment of relative importance of the different reaction steps. An investigation of the contributions to the ozone loss and NO_X budget at different times is performed, see response **A3.4** to reviewer #3.
If we group species O_3, BrO, and $O(^1D)$ and denote the group as O_X, an investigation of the O_X depletion rates for the condition of 200 m boundary layer height can be conducted. The temporal evolutions of O_X depletion rates are plotted in Fig. 3. It is seen that in the early stage (before about day 3.5), the fastest O_X loss reaction is (R31) $HO_2 + O_3 \rightarrow OH + 2\ O_2$. After ozone depletion has started (after day 5), the major O_X loss is caused by the self-reactions (R8) $BrO + BrO \rightarrow 2\ Br$ and (R9) $BrO + BrO \rightarrow Br_2$. It is also noticed that the reaction rate of (R10) $BrO + HO_2 \rightarrow HOBr + O_2$ starts to increase during the induction stage. However, the reaction rate of (R10) is about one order smaller than the rate of (R8) during the ozone depletion stage. Reaction (R10) is more important for O_X at the end of the ozone depletion event. This will be added in the revised paper.
Moreover, the chemical reaction scheme was modified to account for the differences in $O(^1D)$, and the chemical reaction scheme was modified following **R2.7,** see below.

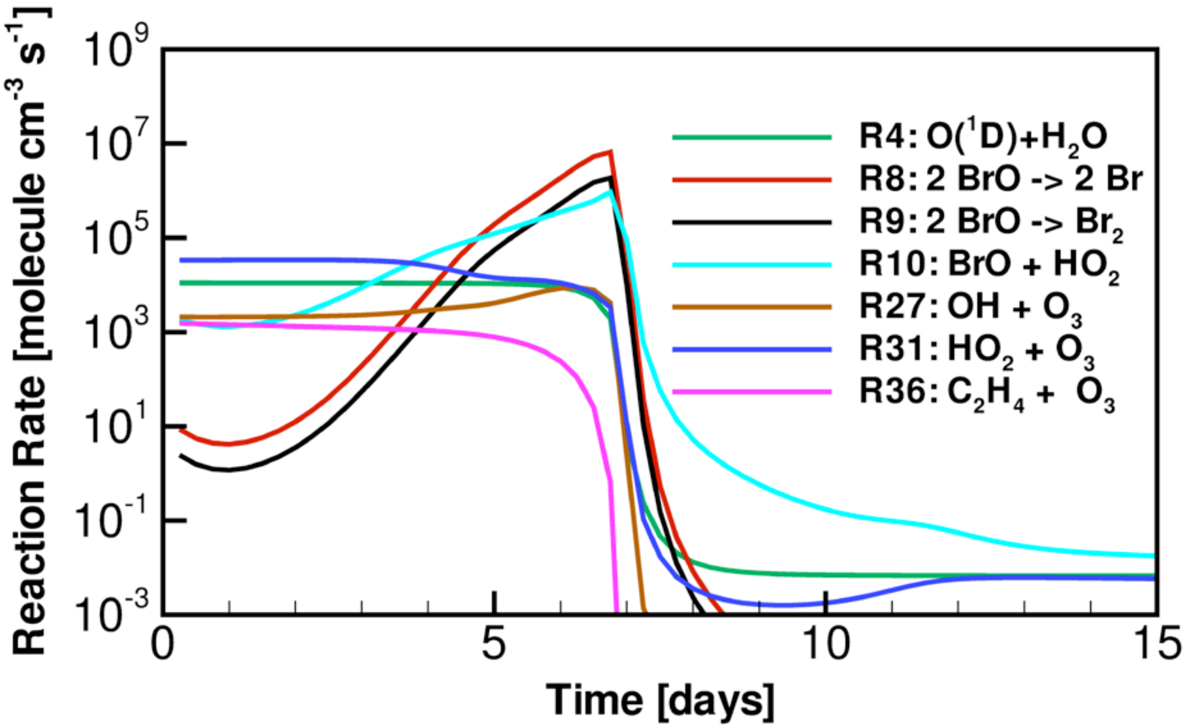

Fig. 3 Temporal behavior of O_X depletion rates in 200 BLH.

R2.6: I suppose that, owing to high mixing ratios (> 30 ppt) simulated for BrO, the self-reaction BrO + BrO (R8 and R9) is a major cycle that leads to a net loss of Ox (= O3 + O + BrO + . . .) at the "depletion stage", rather than BrO + HO2 (R13) (cf. Sander et al., 1997, Fig.2). Statement by the authors also indicates that this is the case (P24184, L21-24). With this argument in mind, implications from the local relative sensitivity matrix (Fig. 4) are very paradoxical in that changes in the rate constants for R8 and R9 do not appear to be a critical factor for the ozone loss in the system simulated here. I encourage the authors to discuss this issue in some details. I also wonder if the simulated HO2 levels are in the right ball park, e.g., around 3 ppt as reported by Mao et al. (2010). On the basis of the calculated sensitivity matrix values, one may even drop R8 and R9 from a skeletal reaction scheme, which the authors plan to construct in future modeling studies (P24185, L1-15). If I understand correctly, such a paradox could be one of the major values of looking at the sensitivity matrices for chemical mechanism reduction in simulating ODEs.

A2.6: We like to thank the reviewer for the valuable suggestion. It is true that the sensitivities of (R8) BrO + BrO → 2Br + O_2 and (R9) BrO + BrO → Br_2 + O_2 are relatively low in the sensitivity matrix which is shown in Fig. 4 in the manuscript. However, it does not mean that they could be dropped **together** in the chemical reaction mechanism. For instance, if (R9) is removed from the mechanism, no significant change will be found in the results of the temporal evolutions of the chemical species mixing ratios. However, the relative importance of (R8) in the new derived mechanism without (R9) will increase a lot, so that we cannot neglect (R8). If we want to continue removing reactions from the revised mechanism, a new sensitivity matrix should be calculated for this mechanism. This discussion will be added in the revised manuscript.

The simulated evolutions of the HO_2 mixing ratio for different chemical reaction mechanisms are shown in Fig. 4, and the value of HO_2 in the bromine only mechanism during ozone depletion is about 0.7 ppt. In the mechanism including NO_X, the value of about 1.2-1.6 ppt is obtained. With the chlorine mechanism, its peak value is about 2 ppt. The values are in the order of the observed value of around 3 ppt, which are mentioned by the reviewer.

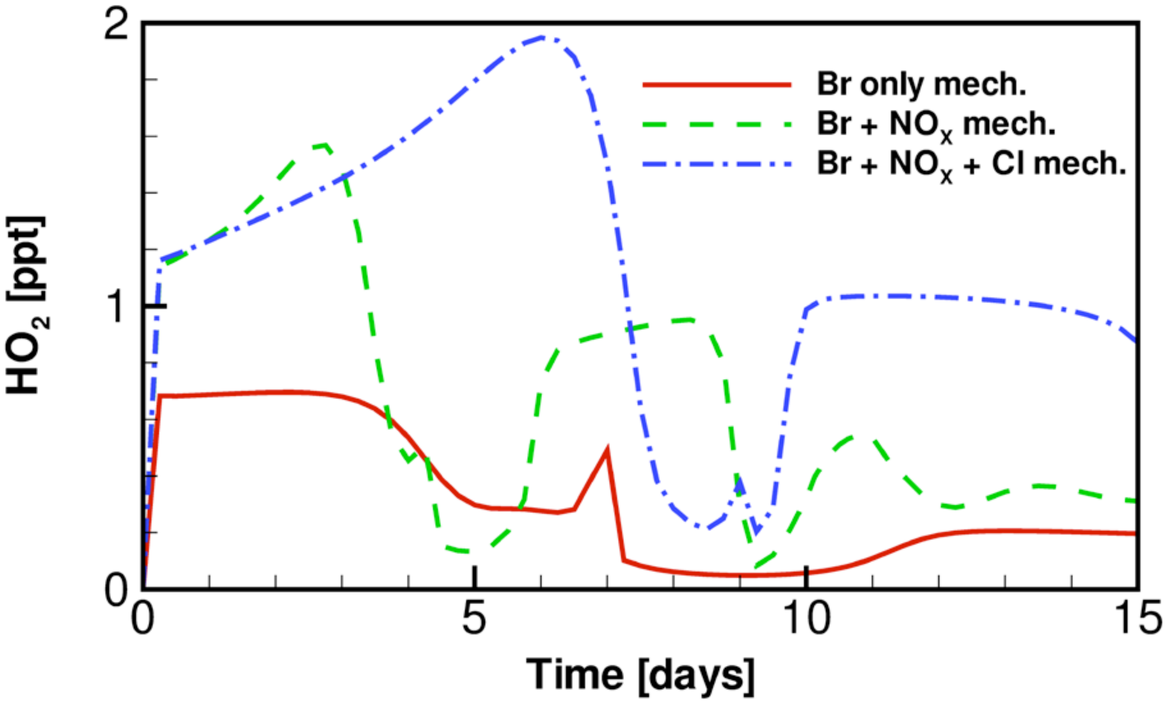

Fig. 4 Temporal variation of HO_2 mixing ratio with 200 m BLH.

R2.7: There is some sloppiness in the chemical mechanism construction. As already raised by Referee #3, “O(1D)” is abbreviated as “O”, which should make most atmospheric chemists puzzled. Also, there are a few missing reactions of potential significance in the polar boundary layer: HONO + hv (sources for both NOx and OH), HCHO + hv (HOx source) and O(1D) + N2 (leading to the suppression of OH formation via H2O + O(1D)). The products of OH + OH (+ M) (R54) should be H2O2 rather than H2O + O3. The assumption that all the C2H5 formed via H-abstraction from C2H6 (R33 & R98) results in the formation of C2H4 + HO2 via R34 (rather than C2H5O2) seems unrealistic. The assumption that the photolytic reaction HNO3 + hv → OH + NO2 is mediated by aerosols and its rate is limited by the uptake of HNO3 from the gas phase (which is quite rapid) seems doubtful. As I see a major value of the paper in its minimalism and the extraction of key metrics, these problems may not be so detrimental as to discredit the entire story. But I do feel uneasy as to how much trust I could place on quantitative terms stated in the paper.

A2.7: The misunderstanding of $O(^1D)$ is corrected. The related reactions are:

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

The product of the reaction OH + OH (+ M) (R54) is corrected to be H_2O_2. We re-ran the code, and it is found that these corrections make hardly any difference for the simulation results.
We disagree with the reviewer in stating that in our mechanism, all C_2H_5 formed results in the formation of C_2H_4, since we also have reaction (R2) $C_2H_5 \rightarrow C_2H_5O_2$, which transforms C_2H_5 into $C_2H_5O_2$. In fact, most of the C_2H_5 molecules in our model are converted to be $C_2H_5O_2$ as the reviewer suggested, since the reaction rate for (R34) $C_2H_5 \rightarrow C_2H_4$ is quite slow. The reaction (R2) was probably overlooked by the reviewer, because its placement within the list of chemical reactions in the supplement is probably not ideal. We will re-order the chemical reactions, and we will move (R2) to the place after (R34).
In the previous simulations for NO_X mechanism, we found that HNO_3 is produced as the final product of the nitrogen related species. However, in order to keep the nitrogen concentration in a reasonable range, we include the multiphase photolysis reaction at the aerosol surfaces to re-activate the nitrogen from HNO_3 (note that the original photolysis is also included as (R67) in the mechanism). However, the photolysis reaction rate at the aerosol surfaces is unknown and difficult to determine. Thus, we use the similar parameterization for this multiphase reaction. However, the parameterization of this reaction can be refined in future.
The authors appreciate that the reviewer identifies possible missing reactions in the mechanism. We found that adding the quenching reaction $O(^1D) + N_2$ will significantly slow down the ozone depletion rate. This might be because $O(^1D)$ is important for the formation of hydroxyl radicals OH, and thus it affects the following HOBr formation. Moreover, the slowdown of $O(^1D)$ formation will affect the concentration of HOBr for the activation processes. In order to clearly identify the role of OH, we added the emission flux H_2O_2 from the ice/snow surface in the bromine only mechanism and increased it to the value 1.0×10^9 molec. cm^{-2} s^{-1}. It is found that this additional flux could overcome the slowing down effect caused by the quenching reaction, as the photolysis of H_2O_2 is a source of the hydroxyl radicals. Moreover, the HONO + hv reaction, as the reviewer suggested, will also offer hydroxyl radicals. However, adding these reactions means that the photolysis reaction rate for HONO and HCHO needs verification, which is left for future research. If the reviewer insists on adding these reactions, we would like to include these reactions and re-run all the simulations and include the results in the revised manuscript if the deadline for revision can be matched.

R2.8: I wonder if the authors could examine the role of BrONO2 hydrolysis from the view-point of simulated NOx concentrations. I encourage the authors to plot NOx versus O3 correlations for different gamma values (cf. Evans et al., 2003, Fig. 5) and see if any insight can be gained into the likely range of gamma(BrONO2)? The same thing could be done for PAN versus O3 correlations (again have a look at Fig. 5 in Evans et al., 2003). Regarding the PAN variability during ODEs, it is also useful to refer to Bottenheim et al. (1986).

A2.8: The modeled evolution of NO_X and PAN concentrations as a function of the O_3 is plotted in Fig. 5 for $\gamma = 0.0001$ and $\gamma = 0.06$, and the results do not show too much of a difference. The mixing ratio of NO_X increases slowly in the early stage and after a steep increase, it decreases rapidly. During the ozone depletion, the mixing ratios of PAN and NO_X keep stable.
Bottenheim et al. (1986) will be cited and discussed in the revised paper.

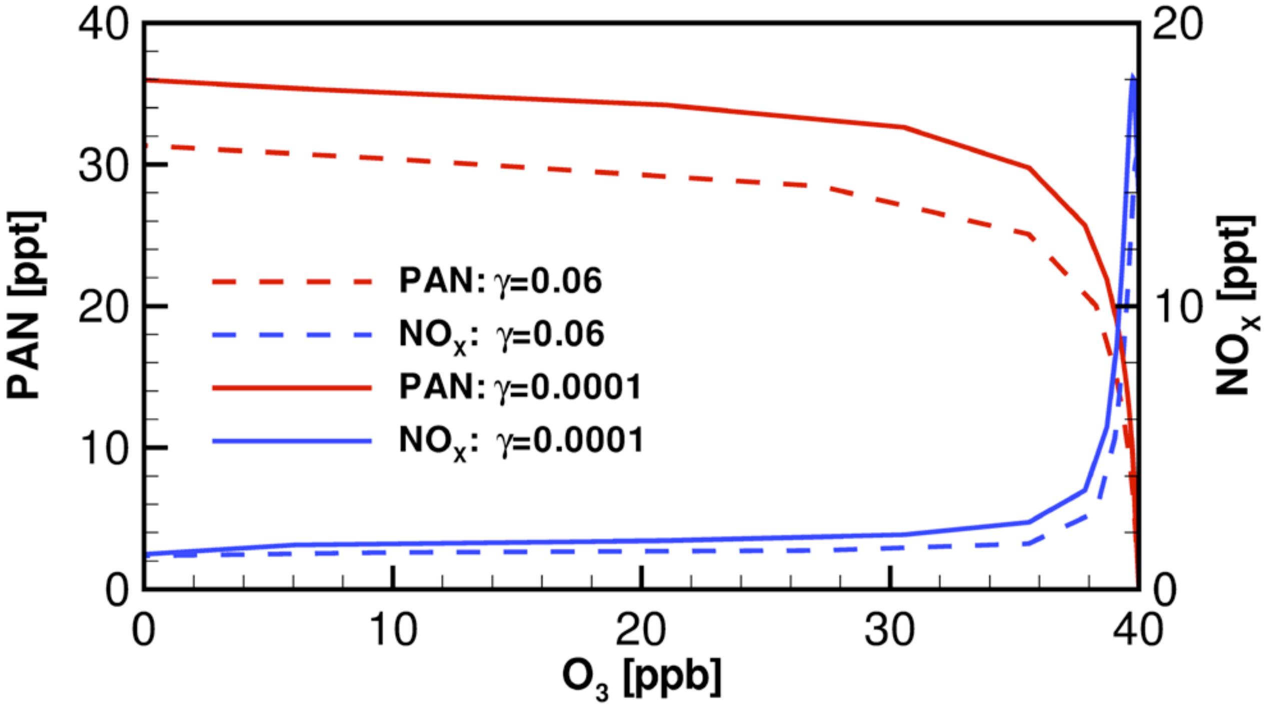

Fig. 5 Chemical evolution of NO_X and PAN mixing ratios during the ozone depletion event in the condition of 200m BLH and γ = 0.0001 and γ = 0.06, plotted against the ozone mixing ratio.

R2.9: As far as I skim through IUPAC and NASA/JPL recommendations, gamma values for BrONO2 hydrolysis appear to be significantly higher than 0.01 whether on the liquid substrate at room temperature (300 K) or on the ice/aerosol surfaces at lower stratospheric temperatures (200 K) but unfortunately without any experimental evidence relevant to the present study (on salty ice surfaces at T 250 K). I do, however, feel that the authors may suggest that the presence of NOx in the polar boundary layer will likely accelerate reactive halogen release and ozone depletion, according to their present results. Please expand the discussion along this line if possible (P24190, L12- 23).

A2.9: The reviewer is right. Based on our model results (the critical γ value obtained for $BrONO_2$ hydrolysis in our model is quite small), we suggested that the presence of this small amount of NO_X in the polar boundary layer (15 ppt in the model) might result in the speed up of the ozone depletion event. The discussion will be extended in the revised manuscript.

R2.10: Discussion in Section 3.3 (chlorine chemistry) seems flawless, but I am not sure if anything new is presented here. I would argue that the authors should move this section to the electronic supplement.

A2.10: We will consider the reviewer's suggestion to move the section to the supplementary material.

Minor Comments:

R2.11: Section 2.2.1: It is largely a repetition from Section 3.2 in Lehrer et al., (2004) and therefore should be shortened significantly. Instead, the authors should give some description of gamma values for BrONO2 here.

A2.11: This section can be shortened, and the calculation details of γ for HOBr may be moved to the supplementary material. The citation of Lehrer et al., (2004) will be mentioned more explicitly.

Section 2.2.1 and 2.2.2 are put just after the description of the heterogeneous reactions HOBr + H^+ + Br^- (or HBr) (P24176, L21 – P24177, L2). Therefore, it might be not so appropriate to put the description of γ values for $BrONO_2$ here – we will put it at an appropriate place in the paper.

R2.12: I find that the parameterization is borrowed largely from Huff and Abbatt (2000, 2002). It should be mentioned more explicitly. Although the impact would not be detrimental, I am not impressed by the inconsistent choice of the values of height (z) and wind speed (u) – by using the adopted formulation, the u value should be taken at the chosen height z, which does not seem to be taken care of seriously by the authors. Also, real-world z0 would change with the wind speed (or friction velocity) as the authors adopted a formulation from König (1985) in their initially submitted manuscript.

A2.12: The citation of Huff and Abbatt (2000, 2002) will be mentioned explicitly in the related context.
About the choice of the wind speed, the value of the wind speed at *z* depends on the geostrophic wind speed, roughness of the surface and the stability of the boundary layer. Considering all these parameters is difficult for a box model study. Therefore, the constant value of geostrophic wind speed is used here.

The value change of z_0 has small effect on the calculation. If we multiply the current value of z_0 by 100 as $z_0 = 10^{-3}$ m, the deposition velocity is 0.78 cm/s, which is not very different from the current value 0.605 cm/s. Thus, we use the constant value of z_0 for the ice surface in the study.

R2.13: P24172, L8: "sea-salt aerosols": The parameterization adopted by the authors inherently excludes halogen release from particulate matters lofted well above the ground level.

A2.13: The words "sea-salt aerosols" will be removed.

R2.14: P24176, L22: Heterogeneous reactions on aerosols (R14, R128) do not change the total loading of halogens in the atmosphere. Hence the authors should modify the sentences around here.

A2.14: The context will be modified as follows:
"In the chemical reaction mechanisms used in this study, the total halogen loading in the atmosphere is determined by the heterogeneous reactions

$$HOBr + H^+ + X^- \rightarrow BrX + H_2O, \qquad (R15, 129)$$

where (R15) refers to X=Br and (R129) to Cl. These heterogeneous reactions occur at the sea ice surface covered by snow and the inert halogen ions are activated from the fresh sea ice, leading to the increase of the halogen mixing ratios in the air. In addition, by absorbing gaseous HOBr and HX, the liquid aerosols could offer a possible location for (R14) and (R128) (Sander and Crutzen, 1996):

$$HOBr + HX \rightarrow BrX + H_2O, \qquad (R14, 128)$$

where (R14) refers to X = Br and (R128) to Cl. In these two reactions, active halogen species BrX are reformed from HX at the aerosol surface, and then, they are emitted into the surrounding environment. Thus, it is essential to precisely parameterize the multiphase reaction rates."

R2.15: P24182, L12-14: Please state values assumed for total column ozone and surface albedo and whether the presence of clouds and aerosols is assumed for photon absorption and multiple scattering in the atmosphere.

A2.15: The photolysis reaction rate calculation in our model is taken from Lehrer et al., (2004), which is evaluated by using a three-coefficient formula with albedo = 1.0:

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

where χ is the solar zenith angle. The coefficients J_0, b, c are determined from the data obtained from Röth's ART model (Röth 1992, 2002) at $\chi = 0°$, 60°and 90°. For the values of the coefficients, please see the answer **A3.1** to reviewer #3. In Röth's ART model, the total column ozone is derived from the initial input ozone profile. The scatterings (Rayleigh scattering) of aerosols and water droplets (Mie scattering) are also included in the model. However, In Lehrer et al., (2004), the initial ozone profile is not provided and no assumptions about the presence of clouds and aerosols are mentioned, so that this question is open.

R2.16: How high is the H2O mixing ratio (or relative humidity) assumed in the model?

A2.16: The value is 0.0008 mol/mol, which will be added in the table of initial values in the revised manuscript.

R2.17: P24182, L18: "CHBr3" should be more appropriate than "CH3Br" to be mentioned here (e.g., Tang and McConnell, 1996).

A2.17: This will be modified in the revised manuscript.

R2.18: P24183, L1: "triggered from the saline surface and sea salt aerosols": I see two problems in this statement: (1) in this sentence, the authors are talking about bromine explosion rather than its trigger, hence they should rephrase from "triggered" to "released", and (2) the parameterization described in Section 2.2.2 implies that the bromine explosion is mediated by salty ice substrate residing very close to the surface level, hence the authors should drop the words "sea-salt aerosols", which are most likely mixed well into the surface boundary layer and above.

A2.18: The context will be modified. (1) "triggered" will be replaced as "released". (2) "sea salt aerosols" will be removed.

R2.19: P24183, L19-20: It is not clear to me how Cao and Gutheil (2011) dealt with bromine release from the surface snow, but I am sure that a 1-D model by Lehrer et al. (2004) implicitly accounted for the effect of aerodynamic resistance imposed on the dry deposition of HOBr from the atmosphere and the release of Br2 and BrCl back to the atmosphere. Hence the authors' statement should be corrected here.

A2.19: The paper of Cao and Gutheil (2011) presents a preliminary study of the chemical reaction mechanism. In that study, the heterogeneous reaction rate at the ice/snow surface is adjusted so that the total bromine concentration at the end of the event equals to the value obtained by Lehrer et al. (2004). Then the comparison of other chemical species mixing ratios is conducted. Although the treatment of the heterogeneous reactions at the ice/snow surface is a little bit artificial in Cao and Gutheil (2011), it proves that the reaction scheme is able to capture the main features of the ozone depletion event (the destroying of ozone and time behavior of the halogen species mixing ratios). The statement will be modified.

R2.20: P24185, L5 & P24211, Fig. 4 (caption): Are the sensitivity values shown here for daily mean or snapshot at a certain time of the day 6? Please clarify. There are other instances where the same vagueness occurs in the manuscript.

A2.20: It shows the instantaneous value at a specified time. This will be clarified in the revised paper.

R2.21: P24192, L20: I would not say that chemical mechanisms used in this study are so "detailed" as compared to models such as Thomas et al. (2011) and Toyota et al. (2013).

A2.21: The word 'detailed' will be relaxed.

R2.22: P24193, L25 to P24194, L2: This paragraph turns up out of the blue and is not substantiated by any part of the present study. I suggest dropping the entire paragraph, as it does nothing in the story line.

A2.22: This paragraph will be removed.

R2.23: P24194, L3-7: Very confusing statement. If you neglect the heterogeneous reactions during the induction stage, what other mechanisms would account for the buildup of halogens in the atmosphere? Please be more specific here. Are you thinking of ad-hoc, 0-th order emissions or something similar? But a sensitivity study with NOx chemistry (Section 3.2) certainly indicates that BrONO2 hydrolysis on the snow/ice surface shortens the induction stage, which seems to me negating the authors' conclusion. I also wonder if the computational benefit of neglecting just a few heterogeneous reactions in the chemical scheme is substantial at all.

A2.23: Even though the heterogeneous reaction rate (e.g., HOBr + H^+ + Br^-) is quite low during the induction stage, 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. This discussion paragraph will be removed.

R2.24: CH3CHO: What is the range of its simulated mixing ratios during the model runs? This species seems to play some significant roles in the present model runs in converting Br atoms to HBr (Fig. 4) and as a precursor of PAN (Section 3.2). These are consistent with what we know could be happening in the Arctic boundary layer, but I would like to see if the simulated CH3CHO mixing ratios change significantly from the initial value of 100 ppt (Table 2). I can see that emissions from the snow are not assumed unlike HCHO (Table 1). From the list of reactions in the supplement, I suppose that CH3CHO is produced photochemically via the degradation of C3H8 (1.2 ppb initially – Table 2) to C2H5O2 and its subsequent reactions. Both of C3H8 and C2H5O2 appear to the surrogate of actual compounds, so the authors are playing a little risky game here.

A2.24: The simulated mixing ratio for CH_3CHO in the model study is shown in Fig. 6 (bromine only mechanism) and Fig. 7 (NO_X including mechanism). They show that the majority of the initial 100 ppt CH_3CHO is converted into HBr during the ozone depletion stage.

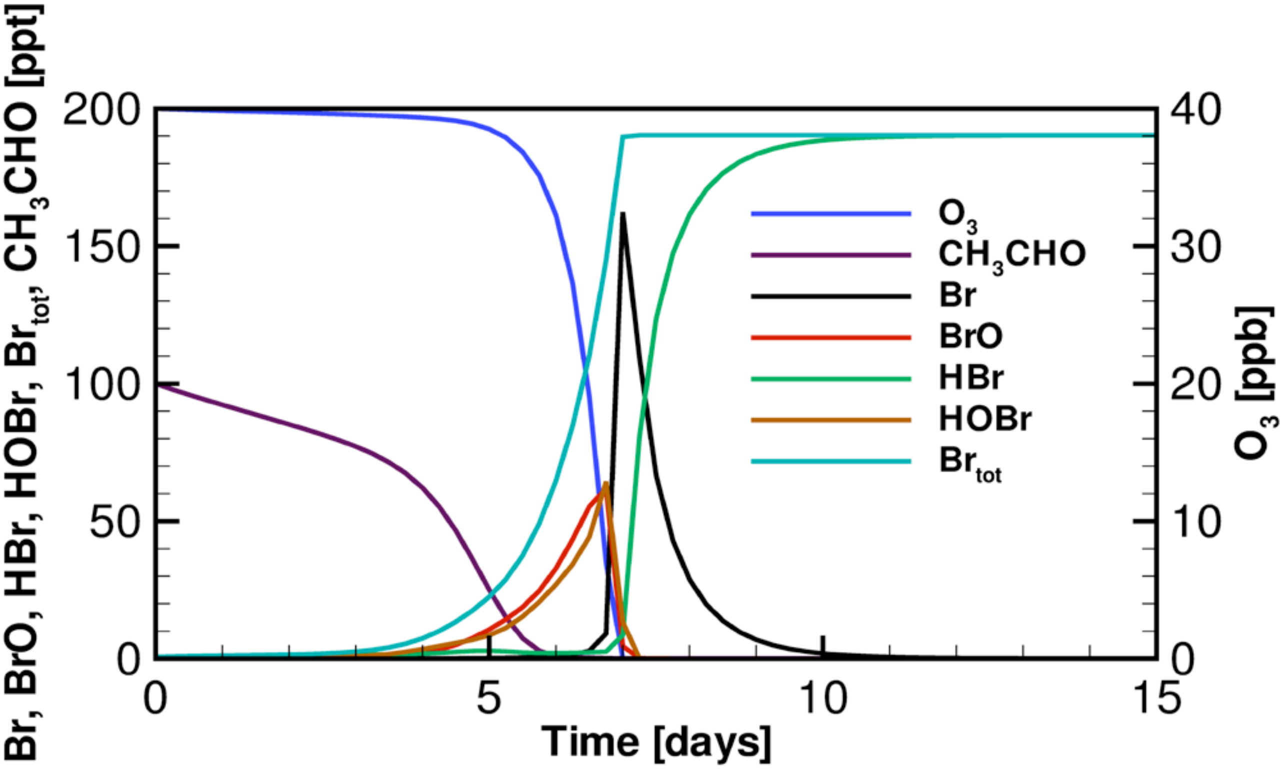

Fig. 6 Temporal behavior of mixing ratios of ozone, bromine and CH_3CHO using the bromine only mechanism for the boundary layer height of 200 m.

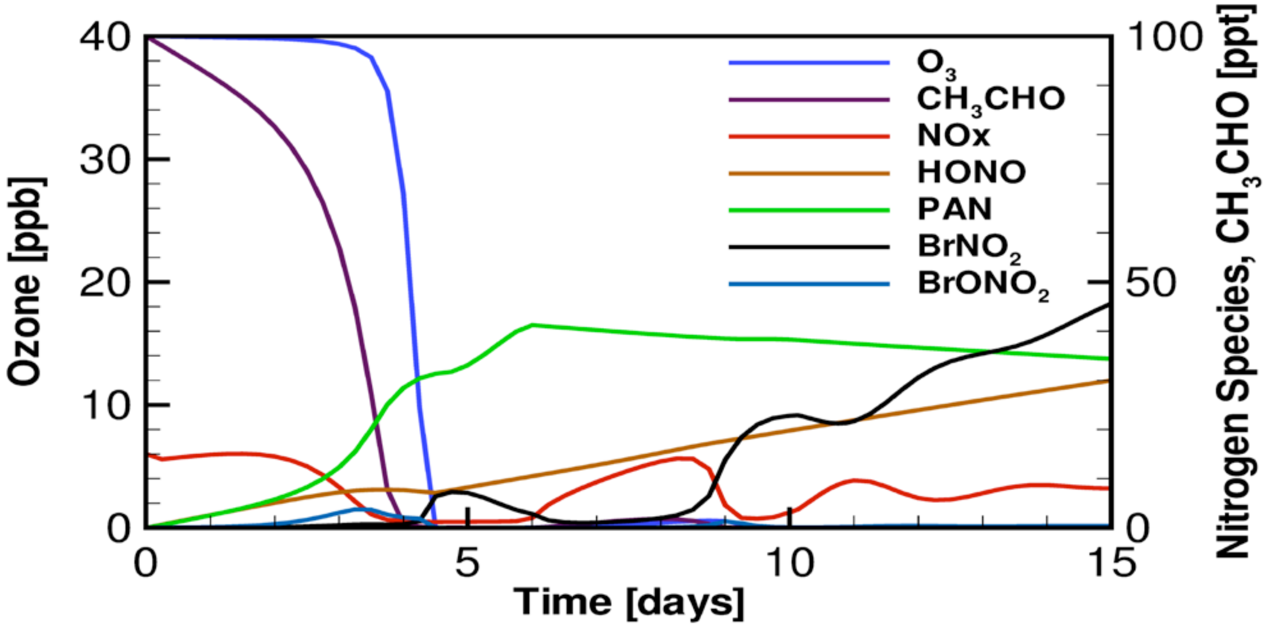

Fig. 7 Temporal behavior of mixing ratios of ozone, nitrogen containing species and CH_3CHO using the NO_X mechanism for the boundary layer height of 200 m.

R2.25: While the present paper is under review, a new paper that follows up the present study appears to have been submitted and accepted in a different journal (Cao, L. and Gutheil, E., Numerical simulation of tropospheric ozone depletion in the polar spring, Air Qual. Atmos. Health, doi:10.1007/s11869-013-0208-9, Received: 31 July 2013, Accepted: 30 September 2013). It would be appropriate if the authors could give a few words to explain how it is related to the present study.

A2.25: In Cao and Gutheil 2013, we performed a 3D simulation using large eddy simulation using a skeletal mechanism, which is derived based on the detailed scheme and the sensitivity presented in the ACP manuscript. It is a follow-up the work shown in the present paper. The description will be added in the revised manuscript.

Editorial Suggestions:

R2.26: Though I see tremendous improvement from the initially submitted manuscript, there remain quite a few loose statements. Wording could be improved further in many places to make it sound more professional for journal publication in atmospheric chemistry. When revising the manuscript, please take an extra caution to editorial aspects again. I have picked up some examples in the editorial suggestions below.

A2.26: The authors appreciate the reviewer's valuable suggestions for the wording improvement of the manuscript. We will revise the manuscript again and improve it.

R2.27: P24172, L2: “In recent years”: This sounds odd, unless your experience in atmospheric chemistry is well beyond 50 years. :-) It has been 25 years since the community recognized the role of bromine chemistry for ozone depletion in the springtime polar boundary layer.

A2.27: The sentence will be changed to “The role of halogen species (e.g. Br, Cl) in the troposphere of polar regions has been investigated for about 25 years.”

R2.28: P24172, L4-5: “auto-catalytic chemical cycle including key self reactions”: If you are talking about HOBr releasing Br2 and/or BrCl from the snow for the former and BrO + BrO destroying ozone for the latter, I think you should phrase so more explicitly here, particularly because the BrO + BrO reaction is not really a part of the auto-catalytic cycle of halogen release.

A2.28: This sentence is replaced as “Halogen species react with ozone to produce halogen oxides followed by an auto-catalytic chemical reaction cycle, leading to the release of Br_2 and BrCl from the sea salt aerosols, fresh sea ice and snow pack.”

R2.29: P27172, L5-6: “several chemical reaction schemes are investigated” → perhaps better by “three sets of reaction schemes are tested by sequentially introducing bromine, nitrogen oxides and chlorine species to the scheme”

A2.29: This will be replaced.

R2.30: P24172, L7: “A category of . . .”: Awkward sentence, but I cannot come up with a suggestion. Please rephrase.

A2.29: We'll do.

R2.31: P24172, L14: "ozone depletion process" → perhaps better by "substantial ozone decrease"

A2.31: We will modify.

R2.32: P24172, L15: "Ozone depletion duration" → perhaps better by "Time required for the major ozone depletion ($[O_3] < X$ ppb) to be achieved" where $X = 1$ or whatever the authors feel appropriate.

A2.32: Ok.

R2.33:
P24172, L16: Drop "a value of".

P24172, L19: "in the order of" → "on the order of"

P24172, L20: "atomic chlorine radicals" → "chlorine atoms"

P24173, L3: "tropospheric layer" → "atmospheric boundary layer"

P24173, L3: "reduce" → "decrease"

P24173, L3: "normal" → "background"

P24173, L14: "bromine concentration in the boundary-layer air"

P24173, L17: "fully" → "thoroughly"

P24173, L19-20: "since the 90's of the last century" → "to date"

P24173, L26: "techniques" → "scenarios" (?)

P24174, L1: ". . . apply the deposition . . . " → ". . . apply the empirically adjusted deposition . . ."

P24174, L24: "confirming" → sounds too strong, perhaps better by "addressing"

P24176, L15: "atmospheric pressure" → "1 atm"

P24176, L21: "halogen stock" → "halogen loading in the atmosphere"

P24182, L11: "radicals" → "atoms"

P24182, L15-16: Reads better by "The initial mixing ratios of gas-phase species are. . ."

A2.33: All the places will be corrected.

R2.34: Unit explanation in the parentheses should move to a caption in Table 2 and then add "ppb = parts per billion".

A2.34: Ok.

R2.35: P24182, L27: Drop “active”.

A2.35: This will be done.

R2.36: P24183, L3-7: Here the words “concentration” and “mole fraction” are used interchangeably, but actually they should not be. If you refer to tracer abundance in the unit of ppb and ppt, you should say “mole fraction” or “mixing ratio”. I would normally choose the word “mixing ratio” and stick to it in the whole sentence.

A2.36: We will stick to the words “mixing ratio” in the revised manuscript.

R2.37: P24183, L12-15: I would rephrase: “When the boundary layer height (Lmix) is raised to 500 m and 1000 m (Figs. 1b-c), the depletion stage commences only after 15 days and even more than 30 days, respectively, and its duration itself is extended to about 3-5 days.” Here I assume that the authors have defined what “induction stage” and “depletion stage” are before this sentence in the revised manuscript.

A2.37: These sentences will be rephrased.

R2.38: P24183, L15-17: I would rephrase: “The speciation profiles of bromine species and their changes with the progress of ozone depletion are similar to those simulated at Lmix = 200 m (Fig. 1a), but the peak concentrations are considerably lower. The maximum values of BrO are in the range of 30–40 ppt, as compared to 60 ppt at Lmix = 200 m.”
A2.38: The sentences will be rephrased.

R2.39: P24184, L4: “keeps stable” → “remains almost constant”

A2.39: Ok, thanks.

R2.40: P24185, L24: “state” → “stage”.

A2.40: Thanks.

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