Rebuttal

The authors like to thank the reviewers for their valuable comments leading to a great improvement of the present submission. We revised the manuscript with modifications marked in red color. We address the comments in detail as follows.

Referee #2

Reviewer:
In this study Herrmann et al. present a 1D modeling study investigating the potential for chemical oscillations (or more generally "recurrences") of ozone depletion episodes (ODEs) in the polar troposphere. Regular recurrences might be expected due to imbalances in the ozone-BrOx equilibrium that is central to the ozone depletion chemistry. For this study the authors used an advanced model involving gas and liquid phase chemistry, gas-aerosol interactions and vertical diffusion between the layers of their model. With initial conditions largely commensurate with observed data during polar ODEs their simulations predict recurrence of ODEs with periods from several days to a month, and ozone recovery from less than 1 to about 10 nmole/mole before depletion restarts. They follow this up with a parameter study to determine the impact of selected parameters in the model on the recurrence of ODEs. The paper is well written with a detailed description of their model, and every simulation result is extensively discussed in the context of the overall ODE mechanism. The main problem with this paper is that by ignoring large scale meteorological effects the model is too simplistic to be relevant.

Authors' Response: We disagree with the reviewer's statement about the relevance of our manuscript. Even though we do not model synoptic meteorological conditions, we are convinced that the results of the model are interesting and important. In the present study, we study whether recurrences are possible without considering large-scale meteorological effects. Therefore, we ignore them in our 1-D model. Also, we noticed that in some field studies, the occurrence of the ozone depletion is interpreted as being caused by the local chemistry (see Jacobi et al., 2006). While we agree that in reality, there will always be a combination of meteorological influences and chemical oscillations, this paper is meant as a contribution to fundamental research on the recurrences of ODEs and a proof of the concept. A perfect simulation including all known physics and chemistry might provide accurate results, but it would also make it very difficult to discern which physical and chemical mechanisms are actually responsible for the oscillations. In a simplified model, it can be tested whether a specific mechanism is responsible for the oscillations. While the results may likely be quantitatively incorrect (in the sense that reality may be different), they still allow to gain a deeper understanding of the specific mechanisms involved.

Reviewer:
There are many definitions on what constitutes a full ODE, but consensus exists that levels of ozone should be $<\sim 10$ nmole/mole, and that the end of an ODE features a return to levels of $>\sim 30$ nmole/mole. Measurements of the rate of ozone loss have been reported that implied total depletion in less than an hour. Satellite data have shown the occurrence of large systems containing enhanced levels of BrOx over the Arctic. All combined this suggests that ODEs as observed are driven by the change of air masses with high ozone/low BrOx and low ozone/high BrOx content, and that the ozone-BrOx chemistry that is responsible for the actual ozone depletion is not what is observed as ODE. Chemical oscillations may well occur during an ODE but are not what drives an ODE: recurrence of ODEs is driven by meteorological variability. Overall, I think the paper is publishable, although the scientific relevance is rather minor. The title should be modified so as not to suggest that the recurrence of ODEs is simulated (maybe into

something along the lines of "simulation study of the oscillations in ozone levels during ODEs").

Authors' Response: We think that there is a misunderstanding. Up to now, the contribution of chemical oscillations to ODEs is not discussed in the literature. The reviewer's statement 'All combined this suggests that ODEs as observed are driven by the change of air masses with high ozone/low BrOx and low ozone/high BrOx content, and that the ozone-BrOx chemistry that is responsible for the actual ozone depletion is not what is observed as ODE.' is not supported by evidence, simply because models including chemical oscillations were not applied to date. While we agree with the reviewer that change of air masses (i.e. meteorology) certainly is important for recurring ODEs, the role of chemical oscillations is simply not known.
We agree, however, that the title should be modified to clarify what the study is about and propose the new title 'On the Contribution of Chemical Oscillations to Ozone Depletion Events in the Polar Spring'. In fact, we are presently extending our previous work (Cao and Gutheil, 2013) using 3D simulations so that the horizontal advection and the vertical convection can be explicitly considered. We have added this information into the last paragraph of the conclusions.

Reviewer:
And while at it I recommend rereading the text and see whether the model description and discussion of simulation results can be made more succinct; the paper is quite long for what it delivers.

Authors' Response: We believe we could save maybe about 1 to 1.5 pages by referring to Cao et al. (2016), however, we prefer to repeat the formulation for an easier understanding of the present model without having to consult other literature. Otherwise, we find it difficult to shorten the text without making the model description or discussion of the results much more difficult to understand.

Referee #3

Reviewer Summary:

Using a 1-D model of photochemistry and vertical diffusion in the lower troposphere, Herrmann et al. examine conditions leading to the self-oscillatory recurrences of ozone depletion events (ODEs) in the modelled polar boundary layer. This study finds that either the vertical diffusion of ozone from the free troposphere or the photochemical production of ozone via NOx chemistry can be a viable mechanism to recover the ozone mixing ratios after each episode of ODE in the boundary layer to drive a subsequent episode of bromine explosion and ozone depletion. Under some model conditions, the system can also approach the steady state after several recurrences of ODE. I have a mixed feeling about the present study. On one hand, the authors have done very thorough model experiments on the emergence and disappearance of the ODE recurrences, expanding a brief report on similar ODE recurrences simulated with a much simpler box model by Evans et al. (2003). On the other hand, the time scales of simulated ODE recurrences are in most cases longer than 10-20 days, whereas the shortest recurrence period simulated is about 5 days with invariant meteorology (turbulent diffusion). The constant meteorology ensures the delineation of the self-regulatory oscillation of the system without external excitation. In the real environment, however, meteorological conditions will certainly vary more or less at this time scale or shorter. Dry (and wet) deposition of aerosols seems to be ignored in the model runs so that particulate bromide accumulating in the model aerosols during the ODEs will stay intact over the recurrence time scales and available for the subsequent bromine explosion event, which I am not sure is realistic enough.

Authors' Response: We are aware of the fact that the aerosols may not stay intact for the entire simulation period. However, as outlined in our answer to major comment 3.) discussed below, the introduction of dry deposition of aerosols to the model does not significantly change the simulation results for the smaller dry deposition velocities. Furthermore, in polar regions aerosols can be also emitted from sources such as frost flowers or blowing snow, so that the loss of aerosols is compensated. The process how the aerosol is transported to the air is still unclear at present. Thus, we feel that it is realistic to consider the aerosols number concentration to be constant. This is made clear in the revised version of the paper, Section 2.1.2 Treatment of aerosols.

Reviewer:
Despite the novelty of this study, I rate its scientific significance somewhere between good and fair, because of the uncertain relevance of self-oscillatory ODE recurrences simulated here. I agree with Referee #2 in their opinion that the title of the paper is somewhat misleading and should be adapted to indicate that the authors have explored the likelihood of the chemically driven recurrence of ODEs in a more or less hypothetical fashion.

Authors' Response: As stated in the second response to Referee #2, we agree that the title should be modified to clarify what the study is about and thus, we propose the new title 'On the Contribution of Chemical Oscillations to Ozone Depletion Events in the Polar Spring'.

Reviewer:
However, who knows if someone will discover from the field data in the future the recurrent feature of ODEs similar to those simulated in this study? Yet it is fair to ask how the varying meteorological conditions can modify the self-oscillatory chemical behaviors. In my opinion, the paper is publishable with relatively minor revisions. It is an interesting paper.

Authors' Response: Thank you for the encouraging comment.

Major comments

Reviewer:
1.) P5, L30-, "To the authors' knowledge, recurrences of ODEs are hardly discussed in the literature": This sounds odd to me. Quite a few studies exist as the basis of our general understanding that variability in the meteorological conditions is the major source of the recurring nature of ODEs. To name a few, in addition to Bottenheim and Chan (2006), Oltmans et al. (2012) reported the role of synoptic air mass transport. Jacobi et al. (2010) pointed out the role of changing local and mesoscale weather conditions including the turbulent diffusion. One may regard model studies by Toyota et al. (2011) and Cao et al. (2016) as the demonstration of meteorological (external) drivers for the occurrence and termination of ODEs. Moore et al. (2014) pointed out the potential role of narrow openings in the sea ice in creating vastly different vertical mass exchange rates between the boundary layer and the free troposphere during the horizontal airmass transport. The authors should rephrase the statement by reflecting on some of such existing studies and should stress that the main value of the present study is in the exploration of other mechanisms potentially causing the ODE recurrences. I also wonder if there is a possibility for resonances with time-varying (periodic) vertical diffusivity profiles, but I guess it will be worth an entirely new study.

Authors' Response: We rephrased the statement to read: 'It is suggested (Hausmann and

Platt, 1994, Tuckermann et al., 1997, Bottenheim and Chan, 2006, Frieß et al., 2011, Oltmans et al., 2012, Helmig et al., 2012) that their cause is transport of air containing varying amounts of reactive Br and O_3 from different locations to the measurement site, leading to recurrence. Jacobi et al. (2010) discussed the role of changing local and mesoscale weather conditions as well as a possibility of a replenishment of ozone via vertical diffusion from aloft. Toyota et al. (2011) demonstrated the occurrence and termination of ODEs by meteorological drivers in a numerical modeling study. Moore et al. (2014) found that narrow openings in the sea ice create vastly different vertical mass exchange rates between the boundary layer and the free troposphere, allowing replenishment of ozone from aloft. Cao et al. (2016) demonstrated in a modeling study the recurrence of an ODE by an instantaneously changing boundary layer structure. Currently unknown is the contribution of chemical oscillations to ODEs, which is the focus of this study.'

We agree that finding resonances with time-varying vertical diffusivity profiles should be possible and interesting to look for, however, that would indeed have to be done in a new study.

Reviewer:

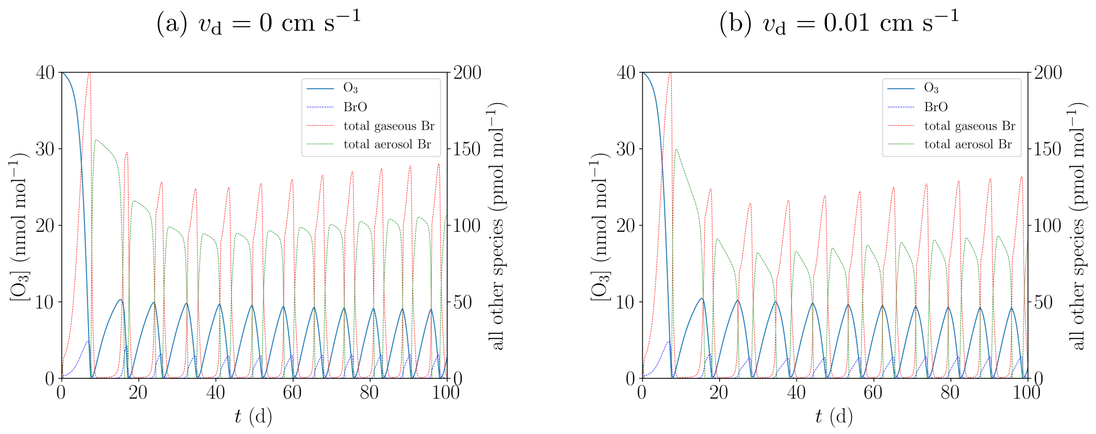
2.) P9, L17, "The pH value is fixed at 5": This assumption is probably good for the pH of moderately acidified sea-salt aerosols (Keene et al., 2002) and NH4-SO4-NO3 aerosols in the NH3-rich environment (Guo et al., 2017), but is perhaps too high for springtime haze aerosols (Li, 1994). Is there a rationale for your assumption and have you explored the role of aerosol acidity in your simulated results? You also mention,"HNO_3 tends to dissolve quickly in aerosols" (P9, L9). If you assumed lower pH values, the behavior of HNO_3 could be different.

Authors' Response: We conducted simulations testing different pH settings. Due to the fixed pH value, there is essentially an infinite amount of H^+ ions. Changing the pH value thus only changes the reaction rate for some of the liquid phase reactions and shifts acid-base equilibria. For a pH value smaller than 7, the recurrence periods do not change. The recycling of bromine for these settings is rate-limited by the aerosol-gas mass transfer, rather than by the aqueous phase reactions. The primary reason for why the bromine recycling is not working at pH values larger than 6, the limited amount of H^+ ions being used up quickly, is still circumvented by the fixed pH. For pH values larger than 7, the ODEs slow down or result in partial ODEs at a pH greater than 9. HNO_3 dissolves slower with decreasing pH, increasing in turn the NO_x mixing ratio for a fixed NO_y initial value. At a pH of approximately 1, HNO_3 mostly stays in the gas phase. It should be noted that the deposition of HNO_3 is overestimated due to the fixed pH, since for a variable pH, the deposition of HNO_3 reduces the pH of the aerosols and thus slows down. Also, HNO_3 is photolysed at the aerosol surface (Grannas et al. 2007), which is currently not implemented in the model. We added the following sentence to the manuscript: 'Simulations found little pH dependence of the oscillation periods for pH values below 7.'

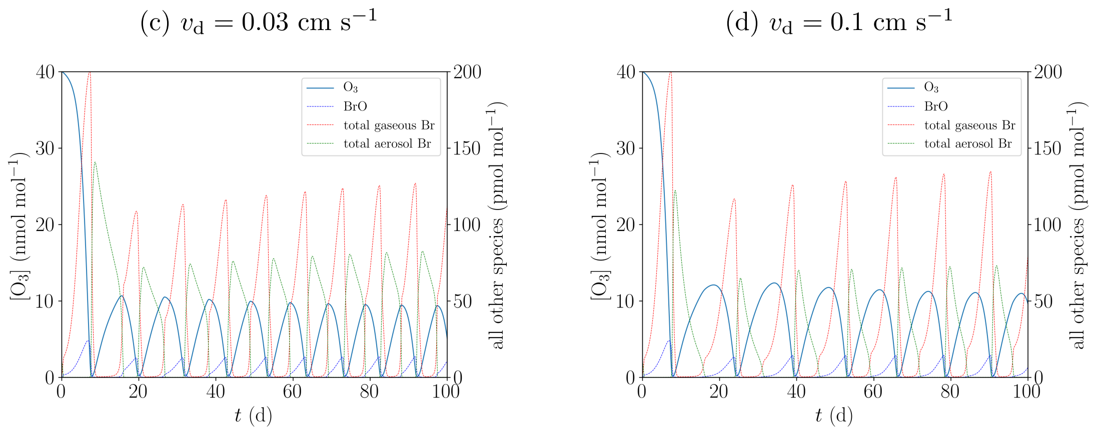
Reviewer:

3.) It seems that the aerosol-phase species are not subject to dry deposition in the present model. Dry deposition velocities of fine aerosol particles are small but not necessarily negligible within the context of this study. If we take the dry deposition velocity of the order of 0.01-0.1 cm/s (e.g., Wu et al., 2018), the residence time of particle-bound species in the atmospheric boundary layer with the thickness of 200 mis estimated to be on the order of 2-20 days. This challenges the validity of neglecting the dry deposition of particle-bound species, in particular, bromide (Br-) (P17, L6-9;C3P19, L2-5). It is probably useful to perform additional model experiments for exploring the impact of dry deposition of aerosols and/or to discuss its implications.

Authors' Response: This is an interesting question. We conducted new simulations including dry deposition of the aerosol phase species Br^-, $HOBr_{aq}$, $Br_{2,aq}$, $BrCl_{aq}$ and HBr_{aq}, employing dry deposition velocities of $v_d = 0.01, 0.03$, and 0.1 cm s^{-1} for the base case settings and different initial values for $[NO_y]$ in steps of 25 pmol mol^{-1} from zero to 100 pmol mol^{-1} NO_y. For 100 pmol mol^{-1} NO_y, the recurrence period increases by approximately 10%, 20% and 40% for $v_d = 0.01, 0.03$ and 0.1 cm s^{-1}, respectively, see Fig. R1. The bromide concentration during the build-up phases decreases by approximately 10%, 20% and 30%, respectively. As a side effect, the ozone peak concentration increases by 3-8% for the smaller velocities and by approximately 20% for $v_d = 0.1$ cm s^{-1}. The NO_y mixing ratio for which the recurrences terminate does not seem to change (about 200 pmol mol^{-1} NO_y for all three cases), and the termination occurs after approximately the same amount of recurrences. However, if we introduce sinks for aerosol species, we should also include sources. New aerosols would likely not start with zero bromide molality. This, however, increases the parameter space further as well as the uncertainties in the implementation of aerosol emissions. These sources may introduce a new way to drive the recurrences. We added some discussion to the revised manuscript on P. 9, L 30-32: "Dry and wet depositions of aerosols as well as productions and emissions of aerosols are neglected. Exploratory simulations show that adding an aerosol deposition velocity in the range of 0.01-0.1 cm s^{-1} (Wu et al., 2018) increases the oscillation period by 10%-40% and decreases the bromide concentration during the build-up phases by approximately 10%-30%. However, if a sink for aerosol is introduced, sources for aerosols such as frost flowers or blowing snow should also be implemented. The produced/emitted aerosols are likely to have non-zero


(a) $v_d = 0$ cm s^{-1}

(b) $v_d = 0.01$ cm s^{-1}


(c) $v_d = 0.03$ cm s^{-1}

(d) $v_d = 0.1$ cm s^{-1}

Figure R1: Simulations for base case settings with $NO_y = 100$ pmol mol^{-1} with different dry deposition velocities for aerosols.

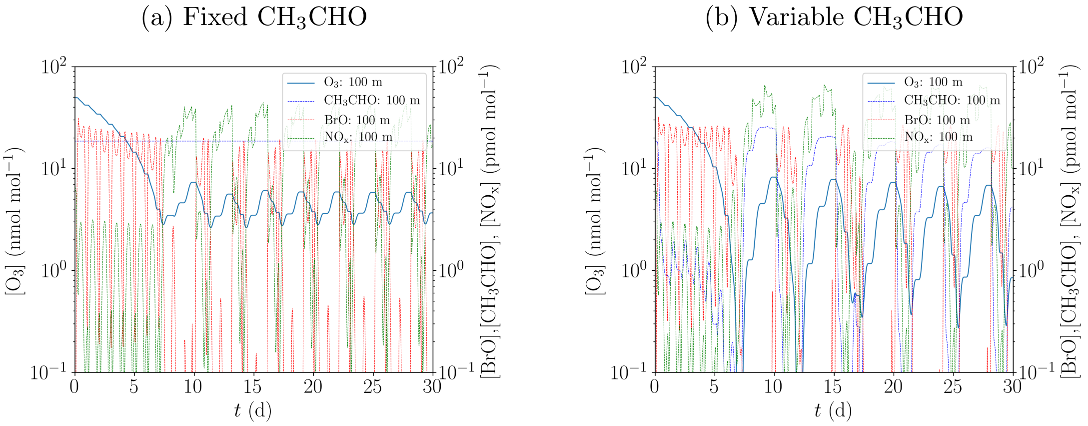
bromide content, providing a source for bromine species and potentially countering the effects of the dry and wet depositions. Therefore, for simplicity and in order to avoid the uncertainties in the production and emission mechanisms of aerosols, both sources and sinks of aerosols are neglected."

Reviewer:
4.) Evans et al. (2003) seem to have assumed the mixing ratio of CH3CHO at 18 pmol/mol (see the end of their paragraph 15), whereas the default initial mixing ratio assumed in the present model is 150 pmol/mol (Table 2). It is not mentioned explicitly whether this value is adjusted for comparison with model results by Evans et al. (2003) in Section 3.3, even though the rate of bromine explosion can be highly sensitive to the CH3CHO mixing ratio. If the simulated mixing ratios of CH3CHO are still on the order of 100 pmol/mol in this case study, it is worthwhile conducting new model runs with CH3CHO = 18 pmol/mol.

Authors' Response: We conducted new simulations with fixed CH_3CHO mixing ratio: The recurrence period does not change at all, however, the first recurrence occurs approximately one day earlier, cf. Fig. R2. The ozone concentration drops only to approximately 2-3 nmol mol^{-1} instead of a mixing ratio smaller than 0.1 nmol mol^{-1}. The reason is probably the reaction of Br with CH_3CHO (R10 in Appendix A1), which produces HBr in the model. The fixed mixing ratio of CH_3CHO converts Br quickly to HBr, terminating the ODE earlier. For the variable case, the mixing ratio of CH_3CHO drops to zero during an ODE, which delays the termination of the ODE; after the ODE, the CH_3CHO concentration recovers to its background level. Evans et al. (2003) took the reaction

$$\mathrm{Br} + \mathrm{CH_3CHO} \longrightarrow \mathrm{HBr} + \mathrm{CH_3C(O)OO}$$

from Minkalowski et al. (2000) with approximately the same rate constant used in the present study. We are not sure why the model of Evans et al. (2003) does not also predict an ozone minimum of 2-3 nmol mol^{-1}. However, at this time, it cannot be determined which value is realistic because of the lack of measurements. Also, the motivation and justification for fixing the CH_3CHO concentration in the study of Evans et al. (2003) remains unclear. In Section 3.3 of the manuscript, it is now stated 'In contrast to the study of Evans et al. (2003), the mixing ratio of acetaldehyde CH_3CHO may evolve freely rather than being kept constant at 18 pmol mol^{-1}.'


(a) Fixed CH_3CHO

(b) Variable CH_3CHO

Figure R2: Simulations of the recurrences of ODEs for the conditions of Evans et al. (2003) for the case with an initial bromide mixing ratio of 43 pmol mol^{-1}.

Reviewer:
5.) Processes that have been investigated in the past model studies but is missing in the present model study include in-snow multiphase photochemistry (e.g., Toyota et al., 2014) as a source of reactive bromine in the atmospheric boundary layer. They may accelerate the build-up of reactive bromine significantly and can thus modify the recurrent behavior of ODEs. This neglected aspect warrants some comments if not tested explicitly by extending the present model experiments.

Authors' Response: We agree that the snow pack may increase the recurrence behavior of ODEs. However, adding a snow pack model with additional physics for the snow pack and its exchange with the gas phase would require major changes to the code. Also, an additional (photo)chemistry mechanism in the snow pack would be necessary. In the conclusions section of the manuscript, P30, L26-29, adding a snow pack model is already suggested as an enhancement of the model in a future work.

Minor comments

Reviewer:
1.) The metric 'α' (defined in Eq. (1)) is referred to when discussing the simulated growth and decay of gaseous reactive bromine. It would be interesting if you can indeed manage to calculate the values of alpha from the model runs and to show their time series along with the mixing ratios of ozone and bromine species. Is it possible at all?

Authors' Response: In general, α is not a well-defined quantity. Considering the bromine explosion mechanism alone, after a time t_0 corresponding to one complete reaction cycle, the bromine mixing ratio has doubled resulting in $\alpha = 2$. The relation

$$[\mathrm{Br}_x](t) = [\mathrm{Br}_x](t=0)\,\alpha^{t/t_0} = [\mathrm{Br}_x](t=0)\,2^{t/t_0}$$

should hold in this simple case. However, if we add other reactions that may deplete or recycle bromine, it is not clear what the time t_0 is and since we can write

$$[\mathrm{Br}_x](t) = [\mathrm{Br}_x](t=0)\,\alpha^{t/t_0} = [\mathrm{Br}_x](t=0)\,e^{\ln(\alpha)t/t_0},$$

we must know t_0 in order to solve for alpha. $[\mathrm{Br}_x]$ is the total mixing ratio of gaseous bromine

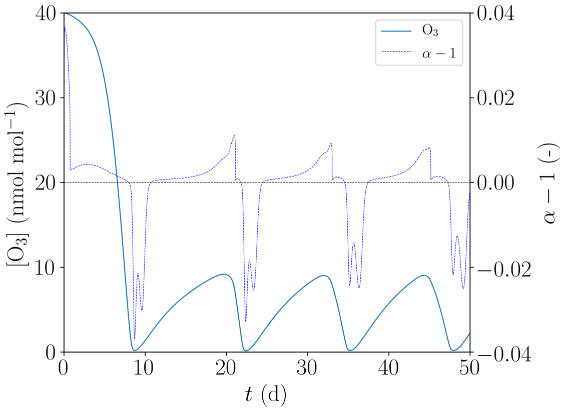


Figure R3: Mixing ratio of ozone and $\alpha - 1$ for base case settings. The dashed horizontal line indicates $\alpha - 1 = 0$

(with Br_2 counting twice). Solving for α and inserting the bromine mixing ratios of two successive time steps with time difference h, we find

$$\alpha = \left(\frac{[\mathrm{Br}_x(t_{n+1})]}{[\mathrm{Br}_x(t_n)]} \right)^{t_0/h} .$$

We estimated t_0 by adding the time scale of the dry deposition of HOBr to the time scales of the two reactions $\mathrm{Br} + \mathrm{O_3} \longrightarrow \mathrm{BrO} + \mathrm{O_2}$ and $\mathrm{BrO} + \mathrm{HO_2} \longrightarrow \mathrm{HOBr} + \mathrm{O_2}$. One might argue that only the slowest time scale should be used instead since the reactions occur simultaneously, however, t_0 is dominated by the slowest time scale in both cases anyway. t_0 is then in the range of 300..1600 seconds depending on the mixing ratio of O_3 and HO_2, with $t_0 \approx 400$ s during an ODE. As shown in Fig. R3, α varies only in the range of 0.96-1.04, where values above unity (i.e. values above the line $\alpha - 1 = 0$ in the figure) indicate bromine explosion and values of α below unity termination of the ODE. The results shown in Fig. R3 imply that already a quite small increase of α above unity is sufficient for the bromine explosion to occur. It should be noted that the value of α varies strongly with the definition of t_0. Also, even though α depends exponentially on t_0, the dependence should not deviate too far from the linear regime since the upper limit of α is 2.

Reviewer:
2.) P6, L17, "...Br instead reacts with HO2, aldehydes or alkenes to form HBr": Models (including the one used in this study) often assume that Br + alkenes produce HBr exclusively, but this is a surrogate approach to simplify complex reactions leading to the production of halogenated VOCs (e.g., Sander et al., 1997; Toyota et al., 2004; Keil and Shepson, 2006). Please consider rephrasing the statement.

Authors' Response: We rephrased the statement on P6, L17. It now reads: "...Br instead reacts with HO_2 or aldehydes to form HBr or with alkenes to form halogenated VOCs (e.g. Sander et al 1997, Toyota et al. 2004, Keil and Shepson, 2006). In many models, including the present formulation, the reactions forming halogenated VOCs are simplified in a surrogate approach to form HBr instead."

Reviewer:
3.) The model description in Section 2.1.1 appears to have significant overlaps with Cao et al. (2016). Please refer to (perhaps minor) changes from Cao et al. (2016) and consider shortening the description if possible.

Authors' Response: We may refer to the section describing the formulation of k_t to Cao et al. (2016) and simply say that we added an inversion layer. 2.1 Moreover, the description of the differential equations might be shortened somewhat. Sections 2.1.2, 2.1.3, 2.2.1, and the following are mostly describing differences to Cao et al. (2016), which cannot be removed. Section 2.4.2 might be shortened by stating that the heterogeneous reactions on snow/ice are described by Cao et al. (2016). Overall, the reference would save about 1 to 1.5 pages, however, we prefer to repeat the formulation for an easier understanding of the present model without having to consult the previous publication.

Reviewer:
4.) From Table A2, I do not see the photolysis of HNO_4 taken into account in this model. According to Stroud et al. (2003): "HNO_4 thermal decomposition and IR photolysis are the important loss mechanisms for HNO_4 in the arctic free troposphere. Our calculations result in

IR photodissociation contributing 20 % and 37 % to the total HNO_4 loss in February and May, respectively." If you believe that this effect can notably change your model results in Section 3.4.2, please discuss.

Authors' Response: We ran new simulations using the HNO_4 photolysis rates of the CAABA/MECCA model. At an SZA of 80°, the photolysis rate of HNO_4 is 7.987×10^{-7} s^{-1} whereas the HNO_4 thermal decomposition at 238K is 7.97×10^{-6} s^{-1}. Thus, the HNO_4 decay is only approximately 10% higher with the added photolysis at that SZA. The results are barely changed by the addition of the photolysis: The NO_x mixing ratio increases by approximately 3% and the recurrence period decreases by approximately 3%.

Reviewer:
5. P26, L5-6: Define what BrNOx represents. BrNO2 + BrONO2?

Authors' Response: Yes, that is what we meant. It is now defined in the text.

Technical suggestions

Reviewer:
P1, L6: as low as → as short as
P1, L13: formulated by Lotka (1909), which are formulated in analogy to → formulated by Lotka (1909) in analogy to
P1, L16: in the order of → of the order of
P4, L25: iodine I (I^- and $IO_3{}^-$) → iodine (I^- and $IO_3{}^-$)
P4, L25: Specify the media (seawater, etc. ?) being referred to concerning the relative abundance of iodine against bromine.
P4, L27: due to the reduction of Cl → due to the reaction of Cl
P9, L14: ...heterogeneous reactions involving NOy ARE FORMULATED TO conserve gas-phase NOy.
P9, L19 and other places: Henry coefficients → Henry's law constants
P10, L4 and other places: uptake coefficient → mass accommodation coefficient
P17, L15: nitrogen oxygen → nitrogen oxides
P19, L2: starting at → restarting from
P19, L3-4: the aerosols are not diffusion limited → the multiphase reactions involving aerosols are not diffusion limited
P19, L4: All other EPISODES start FROM about 30 pmol mol^{-1} of BROMIDE...
P20, L1 and other places: aerosol transfer rates → gas-aerosol mass transfer rates
P23, L5: regenerated → replenished
P24, L13-15 (three times): upper troposphere → free troposphere
P24, L22: transported much MORE SLOWLY
P24, L23: decreasing with larger $k_{\mathrm{t,inv}}$
P24, L24-25: reducing the ozone RECOVERY RATE in the boundary layer and also limiting the maximum LEVELS TO WHICH OZONE CAN BE RECOVERED
P24, L27-: Also, the time between the two recurrences tends to increase PROGRESSIVELY AFTER EACH RECURRENCE since a larger turbulent diffusion coefficient in the inversion layer causes A GREATER loss of bromine to the free troposphere,...
P24, L31: If → In cases where
P26, L26: R59 → R57?
P26, L28: R67 → R65?

P26, L28: R86 → R84?

Authors' Response: We implemented all modifications as suggested by the reviewer.

Reviewer:
P9, L1: It is mentioned that $k_\mathrm{f} = 10$ m^2 s^{-1} here, whereas Figure 1 indicates that $k_\mathrm{f} = 1$ m^2 s^{-1}. Please check the consistency between the two.

Authors' Response: We corrected the caption of Figure 1 of the original paper to clarify that $k_\mathrm{f} = 1$ m^2 s^{-1} for this particular plot.

Reviewer:
P10, Eq. (12): Please double check if the factor "T/T0" is necessary.

Authors' Response: The factor is necessary for the dimensionless Henry coefficients H^cc. If the Henry coefficient H^cc in units [mol liter^{-1} Pa^{-1}] is used, the above factor is not necessary. In the present formulation, the factor T/T_0 arises due to the conversion factor $H^\mathrm{cc} = H^\mathrm{cp}RT$.

Reviewer:
P17, L2-5: You may want to rewrite these two sentences. I do not quite understand the message.

Authors' Response: We rewrote the sentence to read "Both a large ozone regeneration rate and a higher Br release efficiency reduce the drop in the total bromine mixing ratio in the gas as well as in the aerosol phase which occurs between successive oscillations. If the bromine release or the ozone regeneration rate are sufficiently large, the bromine mixing ratio may increase for successive oscillations, as shown in Fig. 7. The additional ozone production due to an increased initial NO_y mixing ratio shortens the oscillation period and therefore limits the bromine loss occurring between successive bromine explosions."

Reviewer:
P24, L23: I cannot see the connection between "drop to approximately 15 nmol mol^{-1}" and "converging to 12 nmol mol^{-1}". Can you rephrase to clarify?

Authors' Response: The '>' sign for 15 nmol mol^{-1} was wrong, it should have been an equal sign '=', thank you. We rewrote the sentence to read 'Ozone is also transported much more slowly to the lower layers of the free troposphere, causing the ozone levels to drop to approximately 15 nmol mol^{-1} at 500 m for $k_\mathrm{t,inv} = 25$ cm^2 s^{-1}; ozone levels decrease further with increased values of $k_\mathrm{t,inv}$, converging to 12 nmol mol^{-1} for $k_\mathrm{t,inv}$ exceeding a value of 50 cm^2 s^{-1}.

Reviewer:
P26, L29-30: I do not quite understand the message here. Do you mean: "The shift from PAN to $\mathrm{HNO_4}$ as the most abundant NO_y species at the lower temperature decreases the net ozone destruction rate during an ODE, resulting in earlier terminations of the ODEs?

Authors' Response: We rephrased the sentence to read "The shift from PAN as the most stable species towards $\mathrm{HNO_4}$ for the lower temperature increases the ozone recovery during an ODE. Since a larger ozone recovery during an ODE facilitates chemical equilibrium with the reactive bromine, this results in earlier terminations of the oscillations of ODEs.'

Reviewer:
P28, L6, "photolyzrefered": Is this a word in German?

Authors' Response: This is a typo, it should read "photolyzed".

References

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