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Atmospheric
Chemistry
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Discussions

Interactive comment on “The potential importance of frost flowers, recycling on snow, and open leads for Ozone Depletion Events” *by* M. Piot and R. von Glasow

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Reply Part 1

We want to thank all three reviewers for their thoughtful and thorough reading of our manuscript and for their comments. We are replying to these comments below and have included the comments for convenience. Our replies are preceeded by ‘Reply:’.

*** Anonymous Referee - #1 ***

p. 2132, l. 4-13: “If we understand correctly, all the calculations do not assume an initial presence of bromine along the trajectory. The surface release is thus constrained by what has been deposited at that instant and takes no cognizance of the fact that it

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would be possible for more bromine to be released [...]"

Reply: See first reply to first reviewer. The reviewer is right mentioning that bromine may be contained in snow along the trajectory. However, the concentration in snow before the "accumulation of events" (mentioned in the reviewer's comment) is completely unclear. As explained p. 4555, l. 21 in our paper, the history of deposition on snow is unknown. We believe the addition of bromine on snow prior to the passage of the studied airmass would add in complexity and uncertainty. Therefore, we intentionally avoided an initial presence of bromine in snow and did not allow the model to release more bromine than is deposited.

"If the surface is an active source, which seems likely then the role of FFs must surely be less important in a instantaneous sense[...]"

Reply: Note that the snow is only considered as recycling medium for deposited bromine. Snow itself is active but not the "real" source. In our model FFs are the source of bromine precursors and snow acts as a recycling surface for the release of reactive bromine. Under our conditions, the role of FF aerosols is important, even in a instantaneous sense.

"1) For the further validation of the FF source function prescribed in the present work it would be useful if the authors emitted Na+ along with Cl- and Br- to the atmosphere and then compare the simulated Na+ and Cl- concentrations in the aerosols and irreversible deposition to the snowpack with any available observations."

Reply: Thanks to the reviewer for proposing this comparison. Na^+ is indeed emitted along with other compounds to the atmosphere. Atmospheric loadings have been compared to measurements from [Hoff and Trivett(1984)] and [Li and Winchester(1989)]. Our modeled concentrations were found to be relatively high (2.0-10.0 μg m^{-3} compared to observations of 0.5-3.0 μg m^{-3}), but note that the history of their sampled air may significantly differ from the air simulated in our model. The ratio [Cl^-]:[Na^+] in snow was found to be similar to observed values [Simpson et al.(2005)]. In addition, we

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compared our irreversible deposition of Na^+ in the model with observed concentrations in snow [Simpson et al.(2005)]. The required deposition period, in order to account for observed amounts in snow, was found to be nearly identical to that for bromine. A note on these comparisons with observations has been added to the text on p. 4555, l. 20.

“2) When discussing the simulated results for the impacts of FF aerosols and FFs themselves on the bromine deposition to the snowpack, the authors assume that snowfall is bromine-free. It would be useful if the authors could put this issue in their context.”

Reply: We agree with the reviewer and have taken this remark into account, p. 4538, l. 18. The following text has been added: “Note that the contribution of snowfall to the deposition of bromide on snow is not taken into account in our model.”

“3): It is interesting to see that the present simulations by the authors never get the surface inversion for about 3 days”

Reply: The model does have a surface inversion which is present during the whole model run. Otherwise, the meteorology would not be correct. The temperature profile shown in Fig. 2 contains an inversion initially at 300 m, but the color chart may not be optimized to see it. We removed the temperature profile and replaced it by the potential temperature profile.

“4) It might be a good idea for the authors to stress the difference between the usual forms of vertical mixing mechanisms such as turbulent diffusion by wind shear (Gong et al., 1997; Strong et al., 2002) and topography-induced disturbance (Morin et al., 2005).”

Reply: The following note has been added to the text, p. 4542, l. 28: “Thus, this dynamical phenomenon is likely to have an influence on the termination of an ODE; also see other possible mechanisms such as topography-induced disturbances [Morin et al.(2005)].”

“The influence of clouds on bromine chemistry in the mid-latitude marine boundary layer was addressed by an earlier study by [von Glasow et al.(2002a)] in which gas-phase bromine species are also taken up by clouds (and aerosols) but only during the nighttime. There is a nice explanation for this behaviour in the paragraph 43 of the von Glasow et al. paper, which is never mentioned in the present paper by Piot and von Glasow. Is it indeed by the different reason that lead to the simulated suppression of bromine radical chemistry in the cloudy Arctic boundary layer? It would be useful to know whether this is the case or not, in terms of difference in the cloud influence between the mid-latitude and polar boundary layers."

Reply: In our paper, the chemical mechanisms leading to the suppression of gas phase halogen in a cloudy environment are similar to those described by [von Glasow et al.(2002a)]. We have added this reference to the paper (p. 4543, l. 21). Thanks for pointing out this omission. In their study, the uptake of halogens into cloud droplets is stronger at night but daytime Br_x and Cl_x also differ from the cloud-free run (see their Fig. 2). In addition to the different LWC, dynamic differences play a role under their as well as our conditions. Also, an important difference with the cloud conditions in our paper is that their cloud cover is permanently present in their model runs.

"5) Lidar measurements detected cloud plumes of buoyant convection from Arctic open leads reaching 3-4 km altitude and it was estimated that open leads as wide as 10 km could produce such intensive plumes during the wintertime [Schnell et al.(1989), Serreze et al.(1992)]. [McElroy et al.(1999)] suggested this possibility for free tropospheric BrO measured in the Arctic spring. On the other hand, the present simulations do not see that level of intensive plumes even for the 1-hour long overpass of open leads (equivalent to 18 km, Runs 4 and 5). We wonder why. Is it because the boundary conditions are totally different? Or, is the MISTRA model intrinsically not capable of handling free convection?"

Reply: As explained in Section 2.2 of the text, a comparison between modeled/measured sensible and latent heat fluxes as well as the turbulent kinetic energy flux [Strunin et al.(1997), Hartmann et al.(1999), Georgiadis et al.(2000), Argentini et al.(2003)] has led to satisfactory results. MISTRA suitably represents the convection over warm and humid open water regions. MISTRA was not developed to model strong (free) convection. The specific conditions in our model runs, however, are such that the upward movement never violates the model assumptions. Furthermore, the lidar measurements from [Schnell et al.(1989)] had been undertaken in winter, which corresponds to quite different meteorological conditions than in spring.

“6) The authors investigated the impacts of NO, NO2, HONO, H2O2, HCHO, and C2H4 emissions from the snowpack individually and concluded that only the HONO emission could exert a notable influence on bromine chemistry (Sect. 3.7). It seems to us that this conclusion is somewhat constrained by their choice of the emission fluxes in the first place.”

Reply: The reviewer is correct pointing out that the emissions from the snowpack are unequally influencing the air chemistry, especially due to their different emission rate values. In this paper we intended to only investigate the presence or not of observed fluxes from the snowpack. A more complete assessment of the influence of these fluxes will be presented in a forthcoming paper. As a consequence, we do not want to include additional information/graphics concerning Sect. 3.7. A note has been added to the text mentioning the publication of a paper in the near future (p. 4556, l. 7).

“If the authors took the HCHO emission estimate of 2.0E+9 molec/cm2/sec from [Grannas et al.(2002)] [...] the present conclusions by the authors might have changed.”

Reply: We note that the value of 2.0×10^9 mentioned by the reviewer was obtained by measurements made in the second half of April (nearly 44 days after Arctic sunrise at Alert). As the photochemistry of HCHO is highly modifying the HCHO atmospheric concentration, we wonder how representative this value is for a study taking place at polar sunrise. A more thorough investigation of the flux of HCHO will be presented in another paper (in preparation).

“Also, the insignificance of C2H4 could have arisen from the MISTRA’s chemical mechanism in which Br + C2H4 produces HBr that is recycled to Br2/BrCl quite rapidly on aerosols. If it is assumed to produce organicallybound bromine compounds that are rather hard to get recycled [Sander et al.(1997), Toyota et al.(2004)], the present conclusion might change. A significant amount (several tens of pmol/mol) of bromoacetaldehyde, which is one of the degradation products from the reaction Br + C2H4, has been detected in-situ near Barrow recently [Keil and Shepson(2006)].”

Reply: The reviewer correctly notes that reaction Br + C_2H_4 may produce organically-bound bromine compounds which are hard to get recycled. Note, however, that concentrations of bromoacetaldehyde given in the paper from [Keil and Shepson(2006)] were not found at several tens of pmol mol^{-1} in April, but rather near detection limit ($\sim$0.5 pmol mol^{-1}). In our model results concerning the influence of C_2H_4 with a flux of 1.3×10^8 from the snowpack, the reaction rate/rate coefficients are too small to account for any significant change on halogen/ozone chemistry. Therefore, producing HBr or more stable organic bromine as reaction products do not matter for the investigated chemistry in this paper. The remark from the reviewer may certainly gain in importance under conditions of high C_2H_4 and reactions producing haloacetaldehyde should be integrated. It will be taken into account in our investigation of the chemistry of C_2H_4 (paper in preparation).

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“Minor comment”:

A) “It appears that sulfate aerosols are present for the recycling of bromine even in non “Arctic-Haze” runs from what I read in Sect. 3.7 (page 4557, lines 10-11). But I miss the aerosol number density and size distribution parameters assumed in those non-haze runs (cf. Table 2)”

Reply: The information is given by: “For a detailed description of the aerosol characteristics, see von Glasow et al. (2002b)”, p. 4530, l. 16-17.

B) “What are the date and latitude conditions for J-value calculations? Also, what time are the sunset and the sunrise in the simulations?”

Reply: This information has been added to the text (p. 4530, l. 3). Photolysis rates are calculated online with the model of [Landgraf and Crutzen(1998)], based on Alert’s latitude (82°N) in the beginning of April (declination of the Sun: +7°). In the model photolysis rates are calculated from nearly 03:25 to 20:35 local time.

C) “Reaction (20), i.e. BrO + HCHO = HOBr + CHO. The rate constant on which the present work is based was derived rather theoretically than experimentally (i.e. via kinetic experiment) by [Hansen et al.(1999)]. The rate constant was derived from a more recent kinetic experiment (although by fitting to complex mechanism) by [Orlando et al.(2000)] and was found to be significantly smaller than Hansen’s. We would like the authors to put a few words about this issue in Sect. 3.2 when discussing the impact of Reaction (20).”

Reply: We thank the reviewer for pointing out more recent calculations of rate coefficient for reaction of BrO with HCHO. We have added the following note to the text, p. 4544, l. 3: “We note, however, that the rate coefficient used in our model is rather an upper limit based on work from [Hansen et al.(1999)]; see more recent studies by [Orlando et al.(2000)]. However, it may be important to notice that calculations by [Orlando et al.(2000)] are also not really adequate as they are only valid for $T=298$ K and do not provide a temperature-dependent function for the coefficient rate.

D) “The CHBr3 deposition is simulated to comprise a non-negligible fraction of the bromine deposition to the snowpack, which is actually a little surprising, but we cannot find its dry deposition velocity in the manuscript and the supplement. How large is it exactly and is there any observational support for it?”

Reply: The gas phase dry deposition velocities are calculated using the resistance model described by [Wesely(1989)]. The dry deposition velocity of $CHBr_3$ in the model is in the range 6-7$\times$10^{-4} m s^{-1}. We are not aware of any observational support which may confirm the validity of this modeled value.

E) “The authors did sensitivity runs to see what happens if nighttime recycling of bromine from the snowpack does not take place but did not definitively conclude that the active nighttime recycling is rather unlikely”.

Reply: Thanks for pointing this out, we have added the following to the text, p. 4547, l. 8: “However, it is important to note that recycling at night compared to daylight recycling is rather unimportant; see observations from [Foster et al.(2001), Spicer et al.(2002)]. Laboratory studies have shown that dark reactions on salty ice may release gas phase bromine [Oum et al.(1998)]. However, their quantitative importance remains to be evaluated.”

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