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Atmospheric
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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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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 - #3 ***

"1A) Why are frost flowers regarded as either a direct source or a sea salt aerosol source? In my opinion, frost flowers are probably both and both at the same time."

Reply: In our paper, we tried to test if FFs do directly release reactive bromine to the gas phase. Within this scope, we chose to model a direct source only, which revealed inconsistency with observations and agreed with conclusions from [Dominé et al.(2005), Simpson et al.(2005), Kalnajs and Avallone(2006)]. If, however, Br_2 is indeed released directly from FFs at the same time with FF aerosols, the bromine explosion would simply accelerate and the ODE would occur earlier. We believe no real quantitative and relevant conclusion can be drawn from such conditions and have not included this case in our paper. Also, we want to stress that no real evidence for reactive bromine have been found over a FF field.

“1A) In model run $\#$ 19 with the direct production of Br2 on the frost flowers the O3 depletion is very efficient. Is that not an indication of the important role frost flowers can play for ODEs? The authors obviously don’t see it this way, because they downplay the results of the model run $\#$ 19 in section 3.8. Why?”

Reply: The O_3 depletion in run $\#$ 19 is similar to the depletion caused in the base run. The Br_2 flux is *prescribed* to induce a MODE4 (similar to base run), see p. 4556, l. 17. It is not surprising to get an efficient ODE via a large production of reactive bromine and this simple representation is not a real indication of the importance of FFs. Our results are not downplayed, they follow observations of no detectable formation of bromine oxide over a FF area [Simpson et al.(2007)].

“1A) Why should the frost flower aerosols, which are obviously solid at the beginning, under these conditions melt or become liquid?”

Reply: We do not think that frost flower aerosols are “obviously” solid. The exact thermodynamics of aerosols released from FFs is not clear as the FF branches contain a quasi-liquid layer of brine. We carefully stated in the text what we considered as FF aerosols: “We assume that the particles released from the quasi-liquid layers present on the FF branches constitute our FF aerosols”. Therefore, these aerosols are considered liquid. Also, see [Koop et al.(2000)] for sea salt mixtures at low temperatures, with temperatures in the model above 240 K (p. 4530, l. 10).

“1A) By the way, such higher sea salt concentrations in the QLL would also apply to the frost flowers still on the ground. Is that taken into account for the model run # 19 with the direct Br2 release from the frost flowers?"

Reply: This representation is only implicitly taken into account in the model run # 19 by prescribing a Br_2 flux. The physico-chemistry of these processes is far from being understood at this point and we mainly wanted to find out in run # 19 how large such a direct release of Br_2 from FF would have to be. A note has been added to the text, p. 4558, l. 12: "(independent of the FF composition)".

"1B) In the model open leads contribute water vapor and sea salt aerosols to the atmosphere and act as an irreversible sink for aerosols. The very strong effect of the increased humidity, the cloud formation and the break-up of the boundary layer structure as described on (P. 4541ff) is very interesting. The authors could stress these results much more."

Reply: We agree with the reviewer. However, we intended to focus on processes which are most often considered important for the chemistry of ozone. Also, to the best of our knowledge, no field observation has correlated the formation of clouds in the PBL to the ozone chemistry yet. We have added the following, p. 4544, l. 27: "The formation of clouds in the PBL may be a significant termination process for ODEs.". We plan to investigate the presence/formation of clouds in relation to ozone in more detail in the future.

"1C) A downward flux of (reactive and less reactive) bromine compounds caused by deposition is calculated. As soon as these molecules touch the surface, 3 out of 4 bromine-containing molecules are (by whatever mechanism) immediately emitted back to the gas phase in the form of Br2 and BrCl. (In some runs the ratio is varied between 0 and 2 out of 4.) Is that correct?

Reply: Yes. Note that the "delayed" re-emission back to the gas phase would not change the chemistry as the FF aerosol release is a (temporary) continuous process.

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“1C) However, the production rate would depend (among other parameters) on the concentrations of bromine compounds (or bromide if you take bromide as the initial source) in the snow, but it will certainly not depend on the downward flux!”

Reply: Note that we assumed no initial bromide concentration in the modeled snow, as we do not know the exact concentration of bromide in snow along a trajectory (see first reply of the first reviewer). In order to calculate the re-emission of bromine from snow, we chose to assume that the chemistry behind the release mechanism is fast enough to immediately re-release part of the deposited bromine to the gas phase. Therefore, the re-release is directly linked to the downward flux as the reviewer wrote himself.

“1C) In summary, this approach is not warranted and is not justified by any measurements I know of.”

Reply: There are very good indications for Br_2/BrCl release from snow. We believe our approach for the Br_2/BrCl is warranted by evidence from, e.g., [Abbatt(1994), Fickert et al.(1999), Huff and Abbatt(2000), Foster et al.(2001), Spicer et al.(2002), Adams et al.(2002)] which show clear production to the gas phase of molecular halogens from solutions containing halides.

“1C) The modeling approach certainly does not represent the processes occurring at the snow surface.”

Reply: We know that we do not model the details, but the reason is that they are not known. What is known is that bromine gets deposited to and released from the snow. As regard to the current understanding of the processes occurring in/on snow, we feel that an attempt to model these processes would only add uncertainties to the calculations.

“1C) I think it suggests that a bromine activation process at the ground would be able (and sufficient) to activate necessary amounts of bromine for an ODE in the entire ABL. Aerosol are not needed in the model.”

Reply: Aerosols are very well needed in the model (as pointed out in the paper), especially for recycling less reactive bromine.

“1C) If that is true and if the snow would provide such a surface, why are ODEs not commonly observed over any given snow surface with a stable ABL?”

Reply: Snow, in our representation, only provides a recycling medium. Snow is not always a source precisely because it is not the “real” source for bromine. Where would bromine in fresh snow come from? Snow may recycle deposited bromine only at places where bromine deposits. See [Simpson et al.(2007)] for more details. The following line has been added to the text to avoid misunderstandings (p. 4532, l. 8): “Therefore, under these conditions snow itself does not represent an initial reservoir of halides and is not a real source of halogens.”

“1C) Or if you put it the other way round: which surface would have the appropriate properties to facilitate a mechanism as incorporated in the model runs? Frost flowers (or more general: new sea ice) comes to my mind”

Reply: We do not attempt to speculate on the most appropriate surface to facilitate a release mechanism. Especially, we wonder if Frost Flowers, as proposed by the reviewer, are indeed appropriate. Again, no detectable reactive bromine has yet been observed over a FF field (see further in the text).

“2) The authors correctly state in their conclusions: “In reality, of course, bromine deposited at night (I would add here “and in the days and nights before”) could be recycled on the next day after sunrise.” (P.462, l. 25-26).” Reply: Done

“2) I fully disagree with the comparison of the model results with the measurements by [Simpson et al.(2005)] regarding the bromine concentrations in the snow as a function of the distance from the open water.”, and “I recommend deleting this paragraph (P.4556, l. 6ff) and also Figure 15”

Reply: We believe this comparison with measurements by [Simpson et al.(2005)] constitutes an important part of our investigation on the potential of snow to recycle bromine. Bromide contained in snow has most likely undergone various deposition processes, which we model by the leap-frogging process. These bromide concentrations in snow are the result of series of accumulation processes. This is what we intend to represent by estimating the accumulated deposition over time in our model via comparisons with measurements by [Simpson et al.(2005)].

“2) Only if it is assumed that the modeled deposition lasts for several days (8 to 24) you get snow concentrations comparable to the measurements.”

Reply: Note that our model runs are in the Lagrangian mode. This means that we only investigated the chemistry of an airmass crossing a source of bromine precursors (in this paper, we used FF aerosols). Therefore, the deposition on snow in our model is the result of one event only. In reality, of course, bromide in snow is, again, the result of numerous deposition events. In order to satisfactorily represent the concentrations of bromide, as measured by [Simpson et al.(2005)], an accumulation period had to be calculated.

“3) Taking into account the above misrepresentations of the processes occurring at frost flower and snow surfaces conclusions like “that bromide activation is unlikely to occur on frost flowers crystals.” (P. 4560, l. 16) or “the best agreement with observations if the recycling of halogens on snow is considered” (P. 4560, l. 10-11) are not justified. These should be deleted. This also affects the title which should be changed accordingly.”

Reply: As we have explained in our replies to the previous comments of this reviewer we do not misrepresent the processes but have improved the explanation of our assumptions better. Therefore we did not delete these conclusions.

“4) I find the figures showing the concentrations and the meteorological data very difficult to read even in the electronic version of the paper. The printed versions are essentially unreadable. This concerns Figures 2 - 4, 6 - 11, and 13.”

Reply: We agree with the reviewer. The quality of the plots has been improved.

P. 4523, l. 25: “This might significantly impact the global tropospheric ozone budget: Any evidence for this statement?”

Reply: We have deleted “significantly” and added [Hollwedel(2005), Simpson et al.(2007)] as reference.

P. 4528, l. 7: “Ludwig-Soret effect: As far as I know the Soret effect has hitherto been applied to mixtures of two neutral organic liquids. I haven’t seen any evidence that it can also be applied to a system of ions dissolved in the QLL of ice. [...] this sentence should be deleted.”

Reply: The Soret effect has been investigated in other mixtures than only two neutral organic liquids. See [Timm and Janek(2005)], and [Kempers(1989)] for multicomponent liquids. However, it is true that it has not been investigated in the specific case of ions in a QLL under Arctic conditions. We base our argument also on [Hafskjold et al.(1993)] theoretically simulating that *the lighter component prefers the hot side of the system at stationary state*. We have modified the sentence p.4528, l. 5-7 to: “Ions contained in the brine migrate toward the crystal branches of the frost flowers through the quasi-liquid layer due to concentration gradients and probably via mechanisms similar to the thermo-migration called Ludwig-Soret effect.”

“If you keep the Zhang et al. paper: it is not correctly cited (issue and page numbers)”

Reply: Done

“P. 4530, l.6: “Photolysis rates are calculated online”: I couldn’t find any further information on the photolysis rates. Maybe I missed it? Are they calculated for a certain date and latitude? What about the diurnal cycle?”

Reply: See “Reply” for minor comment B) from the second reviewer. For more information, the reader is referred to [von Glasow et al.(2002b)].

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“Later on there is the information that in some model runs certain emissions are during the night and so on.”

Reply: It is stated in the text (p. 4534, l. 6-9) that observed emissions are prescribed from the snow in the model. Observed diurnal emissions at field are modeled via binary emission rates. When SZA $> 88^\circ$, emissions are switched off. As ethene does not seem to undergo a diurnal cycle its emission from the snow is continuous.

“[...] Previous measurements of the exchange of H2O2 and HCHO between the atmosphere and snow have demonstrated that the fluxes of both compounds are stronger related to temperature-dependent equilibria than to radiation intensities. [...] so that I would rather expect a flux from the atmosphere to the snow instead of emissions. Since the fluxes do not show any effect according to section 3.7 I recommend repeating the model runs without any fluxes of H2O2 and HCHO.”

Reply: The estimations we did to calculate fluxes of H_2O_2 and HCHO in April are based on comparisons of modeled photolyses between “the period of measurements and springtime” (see p. 4534, l. 10-14). Temperature is implicitly taken into account in this comparison. To the best of our knowledge, springtime measurements of these fluxes are not available yet at locations such as Alert, Canada. Therefore, these estimated values cannot be evaluated with precision. The following sentence has been added, p. 4534, l. 12: “Temperature differences are also taken into account in the comparison of the two periods”.

“P. 4537, l.22: “temperature inversion at around 300 m”: The height of the BL is an important parameter. Unfortunately, it is not clear to me how you determine it”

Reply: The initial conditions are such that the boundary layer height is initially at 300 m. As explained in more detail in [von Glasow et al.(2002b)] we compute the vertical profiles of T, θ, u, v, q prognostically at every timestep. The BL height is defined as the height where the buoyancy term of the momentum flux budget equation exceeds a threshold value (10^{-5} K m s^{-1}). The boundary layer is well mixed up to the temperature inversion as also obvious in the prognostic turbulent exchange coefficients. We feel that the details go beyond the scope of this paper and did not add additional information to the text.

“I also recommend showing the BL height (for example as a black line) in all plots of the temperature and the relative humidity in Figures 2 - 4, 6 - 11, and 13.”

Reply: See “Reply” for the question “What is the initial temperature profile?” asked by the second reviewer.

“P. 4553, l.2: Delete “Due to important climatic implications”: As far as I know “Arctic Haze” has been investigated for several reasons. Better to include an appropriate reference here.”

Reply: We have deleted this sentence.

“Plots of O3 concentrations in Figures 3, 4, 6 - 11, and 13: I suggest adding the 25 nmol mol-1 isopleths to all O3 plots where appropriate.”

Reply: We have added the isopleth to all figures displaying the ozone vertical profile.

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