

Response to Anonymous Referee #2:

General comments

- Cl₂ is constrained to measurements but bromine is not. This makes it difficult to interpret the model results. I would prefer to either constrain all halogens or none.
 - We originally did not constrain the model to Br₂ due to a period of missing data during 29 and 30 March. However, we agree that constraining to the Br₂ measurements, as is done for Cl₂, strengthens this analysis, therefore we adjusted the model to read-in the Br₂ measurements, as well. For the missing period, we used an average diurnal Br₂ cycle from previous days, and then adjusted the Br₂ slightly so that modeled BrO is in line with the observations. An explanation of this has been added to the text on page 15 of the revised manuscript.
- The authors say they could not reproduce the day-to-day variability in Br₂ from only the heterogeneous chemistry. An additional prescribed bromine flux was necessary to estimate Br₂ and BrO observations. Why then is heterogeneous chemistry included in the model at all? Wouldn't it be more straightforward to prescribe gas-phase bromine concentrations directly? What additional value has the heterogeneous chemistry in the model?

The model was constructed to include heterogeneous reactions to investigate whether or not we could achieve the amount of bromine required in the gas phase to agree with observations given known aqueous phase reaction rates and measurements of halide ions in Barrow snow. Since we are constraining the model to Br₂ and Cl₂, the flux of these species from this chemistry is irrelevant. The heterogeneous chemistry does impact the deposition of soluble species (e.g. HOBr, HBr, HCl), and thus their loss from the gas phase through the mass transfer reactions. BrCl is also not prescribed in the model, and the aqueous phase chemistry does contribute to the production of this molecule.
- It is mentioned that the variability seen in the ambient NO_x data is not represented or tested. I wonder if the model results are still useful then. NO_x has a large impact on halogen chemistry.
 - We definitely agree that NO_x has a large impact on the chemistry we are simulating here. Therefore, we have chosen to constrain the model to NO and NO₂ observations, as well, using 10-minute averages of the data. We believe that this change greatly strengthens the analysis as it reflects the NO_x variability. We have left the discussion regarding the NO_x sensitivity of the halogen chemistry limited in this manuscript, however, as this topic is addressed in detail in another paper currently in ACPD: Custard et al., The NO_x dependence of bromine chemistry in the Arctic atmospheric boundary layer, Atmos. Chem. Phys. Discuss., 15, 8329-8360, 2015.
- To increase iodine, the model switches from 1 ppt IO to 0.5 ppt I₂. Why is a different molecule chosen now? I think simply increasing IO by a certain factor would be more useful.

- We used 1 pptv IO and 0.5 pptv I2 because these correspond to two of the very few iodine observations in the Arctic (1 pptv IO by DOAS by Honninger and 0.5 pptv I2 by CIMS by the Shepson group recently). To clarify this, we have changed the terminology to “Low Iodine” and “High Iodine” to distinguish the different modeling scenarios. Figure 3 has now been changed to show the IO levels for the Low Iodine and High Iodine scenarios, and the corresponding I2 levels (0.025 pptv and 0.5 pptv, respectively) are discussed in the text on Page 21 of the revised manuscript.

Specific comments

- Page 28687: “We simulated a 7 day period during late March that included a full ozone depletion event lasting 3 days and subsequent ozone recovery”. The recovery of ozone is mentioned here in the abstract but I cannot find any information in the text how it was modeled.
 - The model was constrained to O3 observations across the 7-day period. This is stated on Page 28694, Lines 9-14 of the original manuscript: "It is important to note that, because our objective was to investigate the halogen chemistry occurring during this time period rather than to attempt to simulate an ozone depletion event, we have constrained our model with the observed ozone mole ratios in order to fully study the fast chemical interactions occurring under these observed conditions, which are a strong function of O₃."
- Page 28691: “Most recently, I2 at levels ≤ 0.5 pptv have been observed by our research group at Barrow, Alaska”. Can you please add a reference to this statement?
 - This reference is currently in submission with the Journal of Geophysical Research (A. R. W. Raso et al., Active Molecular Iodine Snowpack Photochemistry in the Arctic), and we have added this to the manuscript.
- Page 28691: “Halogen atoms can react with formaldehyde” This statement is certainly true for Cl and Br but is the reaction of iodine atoms with formaldehyde significant?
 - Thank you for pointing this out. Reaction with I is not in the model, therefore, we have clarified this statement to read “Bromine and chlorine atoms can react with formaldehyde.”
- Page 28692: “Halogen chemistry also generally increases the Leighton ratio ($[\text{NO}_2]/[\text{NO}]$)”. It should also be discussed here that XO reacts with NO₂ and forms XNO₃. This decreases the Leighton ratio.
 - Yes, thank you for pointing this out. We have added a sentence, on pages 7-8 of the revised manuscript, pointing out this reaction, as well as X + NO₂, and have included these reactions as R17 and R18. However, the net effect of halogen chemistry is an increase in the Leighton ratio.
- Page 28695: What is the difference between J coefficients and J values? If they are identical, why are two different names used?

- The terms “J coefficient” and “J value” were used interchangeably in the text. We have changed all instances to “J coefficient” to be consistent throughout.
- Page 28698/99: “Because the I₂ flux is constant during the ODE, while Br₂, and especially Cl₂ are decreased, iodine becomes more dominant for reaction with O₃ and IO increases.” I understand that the relative importance of iodine increases under these conditions but why should the absolute values of IO increase when ozone is depleted?
 - IO is being produced in the process of ozone depletion, and even though O₃ is low, most I atoms produced react with O₃.
- Page 28702: “it is generally accepted that bromine is the dominant reactant leading to ozone destruction. However, this has not been quantified, nor has this been investigated for varying chemical conditions.” I find this statement too bold. There are many studies which look at this question. I agree, though, that there are still many open questions left.
 - The intent behind this statement was to say that this chemistry has not been examined in detail during very specific chemical conditions, because, prior to OASIS, high time resolution measurements of this large of a suite of species had not been done simultaneously. It was not our intention to imply that no studies have investigated bromine chemistry. We have reworded this statement on page 20 of the revision to read: “However, this has not been quantified over highly variable chemical conditions, since prior to OASIS, simultaneous high-time resolution measurements of such a large suite of chemical species had not been performed.”
- Page 28716: “From our analyses it is clear that the interactions between bromine, chlorine, and iodine are very complex” I think this was already clear before this study started.
 - That is definitely correct, and this was merely used as a transitional statement. We have removed “From our analyses” from this sentence.
- Page 28717: “While chlorine is clearly not necessary to cause ozone depletion, it can significantly impact the rate of ozone depletion.” According to Table 8, chlorine increases the ozone depletion rate from 5.2 to 5.3 ppb/h. I would not call this significant.
 - Thank you, we agree with this assessment. However, after implementing the other changes to the model (e.g., prescribing Br₂ and NO_x), the increase in the ozone depletion caused by Cl is now from 1.85 to 2.94 ppbv/hr. Nevertheless, we have changed this text to read: “While chlorine is clearly not necessary to cause ozone depletion, it can impact the rate of ozone depletion through synergistic effects of cross-reactions that enhance Br atom recycling. Moreover, the presence of elevated chlorine levels can impact important Arctic chemical budgets, including HO_x, NO_x and VOCs, with implications for the oxidative capacity of the PBL.” Thus, we have removed the comparative word “significantly”, and added that Cl chem impacts VOCs, HO_x, and NO_x chemistry.

- Page 287: “In light of these new data, it is crucial that future Arctic modeling studies take into account the activity of chlorine.” Indeed, it is crucial to include chlorine chemistry in the models. However, it is important for hydrocarbons, not for ozone.
 - This is an important point. Given the new numbers for Cl contribution to the O₃ depletion rate (noted above), and the additional sentence we added pointing out that Cl chemistry is important for VOCs, HO_x, and NO_x (also noted above), we have opted to leave this sentence as is.

- Page 28731: The reference Vogt 1999 is incomplete.
 - This has been repaired.

- Page 28740: Why are the heterogeneous reactions of XNO₃ not considered here?
 - We recognize that the heterogeneous reaction scheme for nitrogen oxide compounds is not complete in this manuscript (again this is the subject of Custard et al. ACPD, 2015), however, the heterogeneous reactions of BrNO₃ to produce Br₂ are not a factor here as the model is now constrained to Br₂. This chemistry may have some degree of impact on BrCl, however, the modeled BrCl is within the range of our sparse observations from the campaign, which indicated that BrCl is a relatively unimportant source of Br or Cl. Because our focus here is on the relative impacts of the different halogen radicals, it was important, and a new opportunity, to have accurate relative production rates, as constrained by observations of Br₂.

- Page 28750: In Fig. 4, it seems that the values for Br, Cl, and I add up to 100 %. This is different from the data in Table 7 which is calculated based on equation (4). Why is equation (4) not used for Fig. 4?
 - The reason for the discrepancy is two-fold: one is that the figure is averaged over 3 days, whereas the table is calculated for just midday of one particular day (previously 25 March, now 29 March). Secondly, you are correct that figure is calculated as the fraction by Br, Cl, or I over the sum of the O₃ loss terms, whereas the table used the entire modeled delta O₃ (not just the sum of the terms). To make these consistent, and because O₃ production terms also factor into the delta O₃ (which impact this calculation), we have changed the table to also use the sum of the O₃ loss terms, and have updated equation 4 to reflect this change.

- Page 28750: Another question I have are about the nighttime values in Fig. 4. What is their meaning? At night, the concentrations of ozone-destroying Cl and Br atoms are close to zero.
 - In Figure 4 (now Figure 5), we plot only hours 6 through 21 of the day (from when the sun first rises to when it last sets) and exclude the nighttime values. Even so, you are correct that the absolute values of halogen atoms are very low, however, here we are plotting the relative importance rather than absolute levels.