Authors' response to the reviewer

The authors like to thank the reviewer again for the valuable comments. We revised the manuscript where modifications of the text are marked in blue color. In particular, we address the comments in detail as follows.

Reviewer:
Herrmann et al revised their manuscript describing WRF-Chem simulation of Arctic ODEs and comparison to observations in Utqiagvik, Alaska and Summit, Greenland. The major revision that was completed was the addition of comparison of model results with ground-based observations at Utqiagvik during the OASIS campaign. This was a major undertaking, and the authors are commended for doing this, as this addition significantly strengthened the manuscript, which now adds to the great body of literature stemming from the OASIS campaign. The added comparisons to this data and associated published literature further supports the authors' results, elevating the impact of the results. My comments below focus on the newly added figures and text, with line numbers referring to the tracked changes version of the manuscript.
Figure 4: In response to Reviewer #2 (top of page 14), the authors state that Figure 4 now includes observed BrO (which would be excellent), but I do not see that in either the tracked changes or manuscript file. Perhaps the figure was accidentally not updated?

Authors' Response:

We erroneously wrote that observed BrO is included in Figure 4. We tried to add observed BrO to Figure 4 in an earlier version of the revised manuscript, however, it was extremely difficult to distinguish the lines. As a compromise, we decided to instead add Figure 7 to the paper, which contains only measured and observed BrO and measured $BrNO_2$ for the time range of the observed BrO. We improved Figure 7 of the revised paper, see Figure R1 of this answer. We swapped the colors of measured BrO and simulated $BrNO_2$ and added ozone of simulation 3, which makes a comparison to Figure 4 of the manuscript easier. Also, we changed the aspect ratio of the figure.

Reviewer:
The addition of Figures 7 and S5 and the associated new discussion of simulated production of $BrNO_2$ from reaction of N_2O_5 with Br^- during oil field influence is really interesting and a

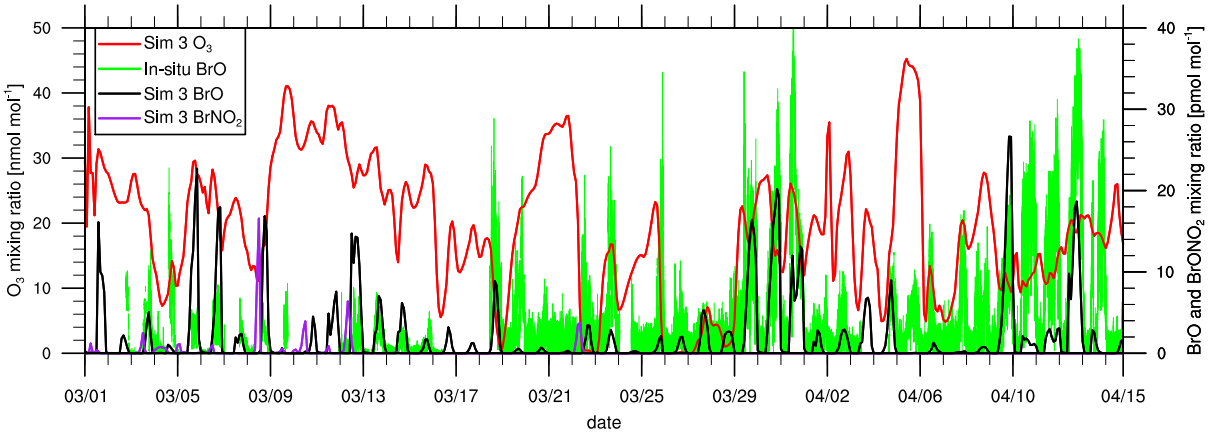

Figure R1: Comparison of modeled BrO and in-situ observations of BrO at Utqiaġvik (Liao et al., 2011) and modeled O_3 and $BrNO_2$; the numerical results are for simulation 3. The date shown is for 00:00, GMT+0.

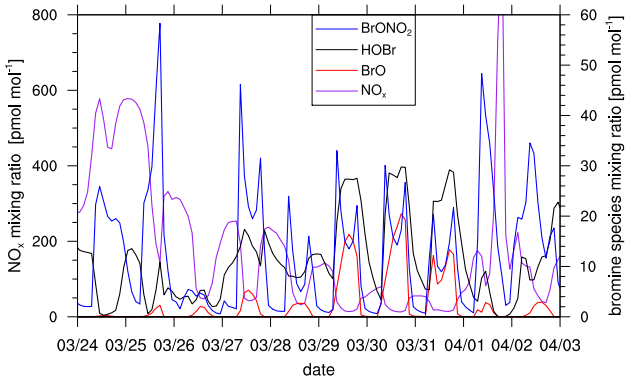

Figure R2: Modeled NO_x, BrO, HOBr and $BrONO_2$ at Utqiaġvik for simulation 3. The date shown is for 00:00, GMT-9 (local time at Utqiaġvik, GMT-9). The time range and timezone is chosen to be directly comparable to Figure 7 of Custard et al., 2015.

great addition to the paper. In particular, higher $BrNO_2$ is predicted earlier during the OASIS campaign when NOx was elevated. This discussion would benefit significantly from integration of discussion of the results of Custard et al (2015, Atmos. Chem. Phys., "The NOx dependence of bromine chemistry in the Arctic atmospheric boundary layer"), who completed 0-D modeling of OASIS, examining the role of NOx in bromine chemistry and predicting BrNO2 production during the same time period as simulated in the current work.

Authors' Response:

Thank you for the suggestion. Figure R2 shows modeled NO_x, BrO, HOBr, and $BrONO_2$ at Utqiaġvik for simulation 3, which may be compared to the results of Custard et al. (2015). Figure R2 is added to the supplement, as a subfigure of S6. The following discussion is added to line 400 of the revised paper: 'Custard et al. (2015) studied the role of NOx in bromine chemistry from the March 24, 2009 to April 3, 2009 at Utqiaġvik using a box model. They found a suppression of ozone destruction for a high NO_x case (concentrations in the range of 800 to 1600 pmol/mol). During this time frame, the simulation with WRF-Chem predicts negligible production of reactive bromine due to N_2O_5. In Fig. S6 of the supplement, modeled Modeled NO_x, $BrONO_2$, HOBr and BrO is shown for the time range modeled in Custard et al. (2015). Modeled NO_x is elevated from March 24 to March 26 and again on April 2, similar to the measurements of NO_x shown in Fig. 2 of the paper of Custard et al. (2015). However, the present model does not find NO_x mixing ratios on the order of 10,000 pmol/mol as found on March 24-27 in the measurements. The typical modeled NOx concentrations are in the range of 50 to 1000 pmol mol^{-1}, i.e. between the high and low NO_x scenarios of Custard et al., 2015. The predicted values of $BrONO_2$ compare quite well with these of Custard et al. (2015), see Fig. 7c of that work, with peak values around 50 pmol/mol.'

Reviewer:

Several places in the manuscript refer to an under-prediction of BrO over land, and it is stated that this is discussed in a later section (presumably the brief mention on page 28?). To support this, I suggest adding a sentence on Line 509 that refers to Pratt et al (2013, Nat. Geo.) and Peterson et al. (2018, ACS Earth & Space Chem), both of which report MAX-DOAS BrO observations over the tundra snowpack (up to >100 km inland). In fact, in the Utqiagvik region, Peterson et al (2018) observed higher BrO over the tundra than the FYI. Adding a sentence referring to previous measurement of BrO over inland tundra snowpack provides an explanation for the current study's result (given the short lifetime of BrO) and will strengthen the manuscript as a result.

Authors' Response:

We added the following text to line 492 of the revised manuscript: 'Pratt et al. (2013) and Peterson et al. (2018) reported BrO observations using MAX-DOAS over the tundra snowpack, which show elevated BrO levels up to more than 100 km inland. Peterson et al. (2018) found higher BrO concentrations over the tundra than over FY ice. In contrast to that, the simulations conducted in this work under-predict BrO over land...'

Additional comments

Reviewer:
Line 2: In response to Reviewer #1, the authors changed "studied using the regional software WRF-Chem" to "studied using the open-source software package WRF-Chem". However, this didn't address the reviewer's comment. It is important for someone not familiar with WRF-Chem to understand that it is a regional model, and not another type of software.

Authors' Response:
Thank your for the suggestion, we agree that it is important to mention the usage of a regional model. In the revised paper, line 2 reads '...are studied using a regional model based on the open-source software package WRF-Chem.'

Reviewer:
Line 4: In response to Reviewer #1, the authors clarified elsewhere that Br2 is emitted from snow above sea ice rather than the sea ice itself, but this sentence still needs to be updated by deleting "ice and". Also, Lines 577-578 needs to be revised as well, as it states "...oxidation of bromide by ozone directly from the sea ice."

Authors' Response:
We removed the words 'ice and' from line 4. Lines 577-578 of the revised manuscript now states 'Bromine may be emitted by the extended bromine explosion mechanism and/or oxidation of bromide by ozone directly from the snow covering sea ice.'

Reviewer:
Line 25: I suggest changing "is most likely destroyed" to "is destroyed".

Authors' Response:
We changed line 25 as suggested.

Reviewer:
Line 49: I suggest adding the following to this new sentence "...field-based experiments, and Wren et al. (2013, ACP) and Halfacre et al. (2019, ACP) through lab-based experiments."

Authors' Response:
We modified line 49 as suggested.

Reviewer:
Lines 214-215: I suggest replacing Wang et al. (2019b) here with the more appropriate Halfacre

et al. (2019, ACP).

Authors' Response:
We reworded the sentence slightly and changed the reference as suggested.

Reviewer:
Figure 7: The addition of this figure is quite valuable. However, it is very difficult to discern the black trace as currently plotted. Also, the time zone plotted needs to be stated, either in the figure or caption, in this figure and all other similar figures (Figures 2, 4, 10, 11, 16). I also suggest removing the "00:00" from the x axis labels and instead label more ticks to make it easier to interpret by having more dates labeled.

Authors' Response:
The time zone is now stated in the captions of the Figures in the revised manuscript. Also, we improved the Figures as suggested. Figure R1 shows the revised version of Figure 7 of the manuscript.

Reviewer:
Figures 6, 8, & 9: Please provide the time zone that the vertical profile times correspond to.

Authors' Response:
The time zone now is mentioned in the caption of Figures 6, 8, and 9.

Reviewer:
Figure 9 bottom-right: If the BrO observations above 100 m are known to be inaccurate as stated in the caption, then they should not be plotted in the figure.

Authors' Response:
We changed the figure as suggested, see Figure R3, which replaces Fig. 9 in the manuscript.

Reviewer:
Lines 441-442: Reference to Moore et al. (2014, Nature) should accompany the added text here.

Authors' Response:
We added the reference to Moore et al. (2014, Nature) to the suggested lines.

Reviewer:
Line 508: Rather than citing Pratt et al. (2013) here, I suggest adding Jacobi et al. (2012, JGR, "Chemical composition of the snowpack during the OASIS spring campaign 2009 at Barrow, Alaska) since it describes the snow composition during the OASIS campaign, and Krnavek et al. (2012, Atmos. Environ., "The chemical composition of surface snow in the Arctic: Examining marine, terrestrial, and atmospheric influences"), since it compares [Br-] over tundra, FYI, and MYI, to the publications already listed. I encourage the authors to review these and other Arctic snow composition measurements studies, which refute their statement on page 18 of the response that "the assumption of no bromide content of snow on land or near coasts can be correct in many circumstances", as this statement is not supported by measurements.

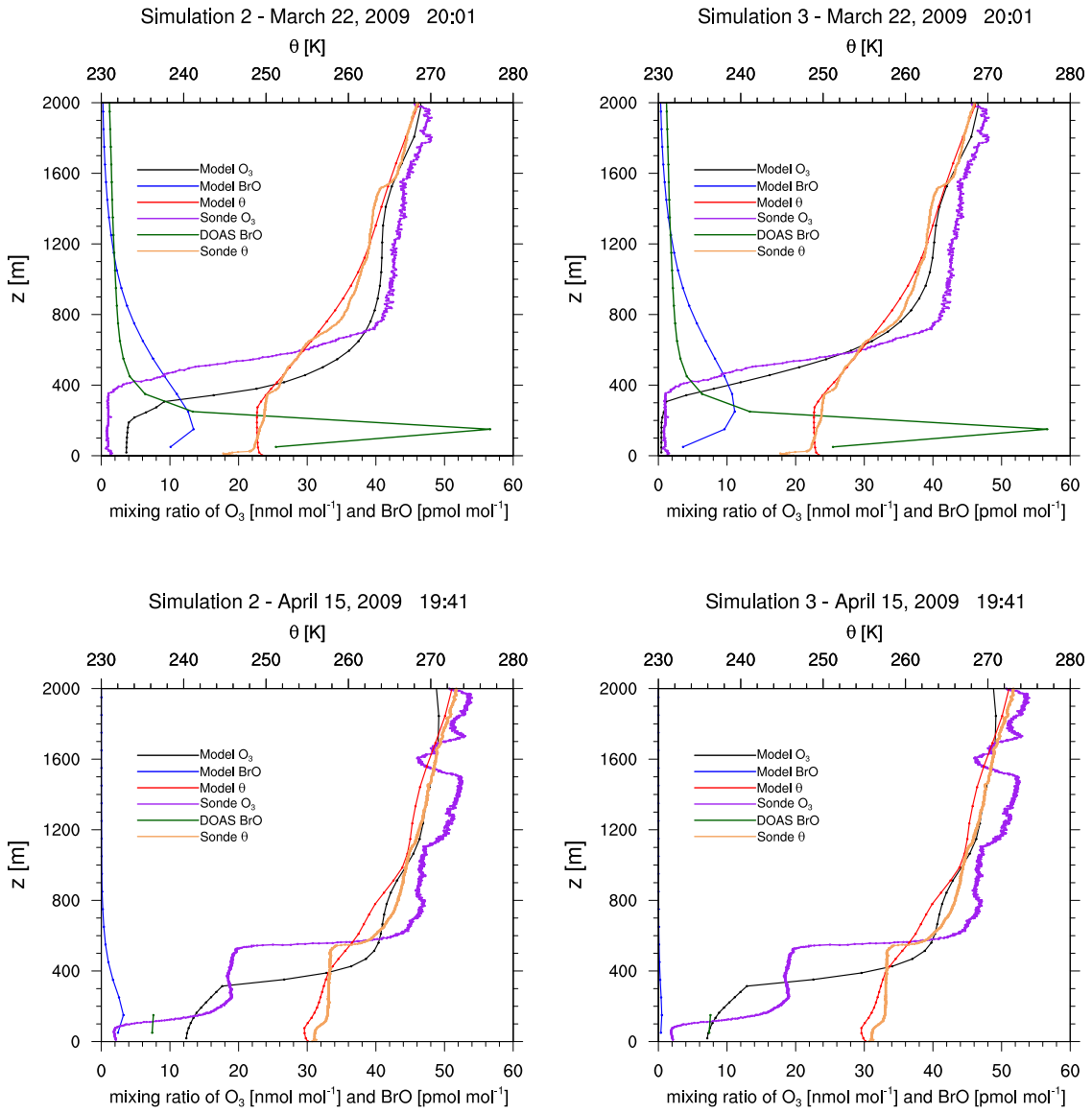

Figure R3: Vertical profiles of measured and modeled (simulation 2 (left) and simulation 3 (right)) ozone, of potential temperature θ, and of BrO at Utqiaġvik on March 22 (top) and April 15 (bottom), 2009. The time zone is GMT+0. Measurements are from upward flights using ozone sondes (Oltmans et al., 2012) and DOAS measurements (Frieß et al, 2012). On April 15, only the observed BrO mixing ratio in the lowest 100 m is accurate due to very poor visibility.

Authors' Response:
We added Jacobi et al. (2012) and Krnavek et al. (2012) to Line 508 of the revised manuscript, as suggested. We are aware of the fact that the bromide content of snow can be non-zero. However, with regard to bromine emissions from the snow, it is sufficient for the bromide content to be sufficiently small for the assumption of no bromide content to be approximately correct. In the manuscript we write: 'The assumption of zero bromide content of snow covering land or MY ice is of course an idealization and not always correct in reality (Simpson et al., 2005; Jacobi et al., 2012; Krnavek et al., 2012; Peterson et al., 2018, 2019), contributing to the underprediction of BrO over land mentioned in this paragraph.' We note that the measurements of snow bromide span a range of nearly three orders of magnitude (Jacobie et al., 2012, find 5 to 1400 μg/l, depending of the type of snow).

Reviewer:
Response Page 16: Note that Wang and Pratt (2017) simply used jBr2 as a term to define the timing of radiation-dependent emission of Br2 from the snowpack as a representation of the observations by Pratt et al. (2013), which showed the Br2 was produced from Arctic snow in a chamber only upon irradiation and no addition of O3 or other gas-phase oxidant. This mechanism was replicated in the lab by Halfacre et al. (2019, ACP), who showed Br2 production upon irradiation of ice containing Br- and an OH precursor. Therefore, it is incorrect that Br2 is required for condensed-phase snowpack Br2 production.

Authors' Response:
We are sorry for the misunderstanding. In Wang and Pratt (2017), section 3.2, two different parameterizations of snowpack emissions are described:
'(i) JScale: emission rates (FX2) are scaled linearly with j-values (i.e., $F_{X2} \propto j_{X2}$, where X2 = Cl_2 or Br_2);'
'(ii) SS: emission rates are scaled with steady-state (SS) removal (i.e., $F_{X2} \propto j_{X2} \cdot [X2]$)). The SS parameterization is based on the steady-state assumption that the snowpack emission rates of Br_2 and Cl_2 are balanced by their photolysis, due to the short daytime photolysis lifetimes of Br_2 and Cl_2 in the Arctic (tens of seconds and tens of minutes, respectively, for March 2009 in Utqiaġvik;'
We tried to address both parameterizations in our response, saying that parameterization (ii) should be, in our understanding, not suitable to describe the initiation of a bromine explosion, since the parameterization assumes the emission of reactive bromine to be proportional to the concentration of Br_2. Therefore, at zero Br it will produce a zero Br-flux and thus a Br-explosion can never start. Parameterization (i), which is addressed later in the same response on page 16, of course does not require Br_2, as the reviewer state, and thus can lead to the start of a Br-explosion. The manuscript was not changed in response to this question.

Reviewer:
Line 623-624: I encourage the authors to add acknowledgement of the individuals that conducted the OASIS measurements and produced the data used. These BrO measurements by CIMS and DOAS are not trivial whatsoever, and these individuals should at least be recognized here, as their data significantly contributed to the manuscript. While H. Sihler and U. Platt are listed under the author contributions, there were other individuals that conducted the CIMS measurements, in particular.

Authors' Response:
Thank you, in the revised manuscript, we thank J. L. Liao, L.G. Huey and D. J. Tanner, who conducted the CIMS measurements: 'The authors thank J. Liao, L. G. Huey and D. J. Tanner,

who conducted CIMS measurements during the OASIS campaign.'. U. Frieß and H. Sihler, who conducted the DOAS measurements, are co-authors of the present paper.

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