Authors' response to Reviewer 1's comments

We thank the reviewer for the helpful and constructive comments. Our responses to the reviewer's comments are included below following each reviewer's comment. The reviewer's comments are shown in *italic*, and our responses are in blue.

This manuscript describes a modelling study of Arctic tropospheric ozone using two chemical transport models (DEHM and GEM-MACH), each parameterized with a different bromine source mechanism (blowing snow and snowpack, respectively). Notably the regional models were run at high spatial resolution (~20 km). Sensitivity tests were performed for bromine chemistry and boreal wildfire emissions. This is an important modelling study that includes comparisons to a large number of observational datasets - not only ground-based monitoring sites, but also buoys, mobile platforms, and vertical profiles. I expect this is likely the most comprehensive high resolution model-measurement intercomparison thus far. The manuscript is comprehensive and well-written, with clear figures that enhance understanding. The authors do an excellent job of comprehensively citing literature and discussing their results throughout the paper. Specific comments below mainly focus on improving clarity.

DEHM Methods: How was blowing snow itself parameterized? I did not see that described in the methods. Chen et al. 2022 (ACP, https://doi.org/10.5194/acp-22-15263-2022) showed that the commonly used parameterization based on wind speed and temperature overpredicts blowing snow conditions, likely due to the lack of inclusion of a snow age term.

R: Thank you for this comment. DEHM parameterizations of the source term of Bromine from blowing snow are based on the parameterization described in Yang et al. (2008) including the temperature dependent threshold for the lifting of snow and the snow-age dependent attenuation factor A' which significantly reduce the lifting within the first 24 hours after a snowfall event. The age of the snow is calculated as number of hours since the last snowfall event based on WRF model output of hourly accumulated snow field. It does not consider horizontally transported snow from one grid cell to another, which could also influence the age of the snow. We have added the following (**in bold**) in the original text:

"For blowing-snow production of sea salt, Yang et al. (2008, 2010) made use of a blowing snow sublimation rate, which is a complex function of wind speed (at 10m), air temperature, relative humidity, snow age, etc. **For the implementation in DEHM, the calculation of the temperature-dependant wind speed threshold for lifting snow and the attenuation factor, which reduces the lifting of snow as a function of the age of snow, are the same as described in Yang et al. (2008). The age of the snow is estimated as the number of hours since last snowfall events in the WRF model output of hourly accumulated snow fields. It does not consider horizontally transported snow from one grid cell to another, which could also change the age of the surface snow.** For this study, the size dependent salinity of snow in Yang et al. (2008) was scaled to a mean salinity for the Arctic of 0.93 psu for snow on FY sea ice, …"

Given the stated goal to investigate key processes driving surface O3 seasonal cycles, combustion NOx emissions have recently been shown to have regional impacts on bromine recycling and ozone, as described by Peterson et al. 2025 (Faraday Discussions, https://doi.org/10.1039/D4FD00166D) and Widmaier et al. 2025 (Faraday Discussions, https://doi.org/10.1039/D4FD00166D). Since this wasn't investigated in the current study, the authors are encouraged to at least add a sentence in the conclusions pointing to this as a suggested future direction.

R: We appreciate that the reviewer brought to our attention the recent studies highlighting the role of anthropogenic NOx emissions (e.g., Northern Alaskan oil fields near Utqiagvik) in bromine and O3 chemistry influencing surface O3 seasonal cycles on both local and regional scales. Although some of the results from our current study did reflect the individual effects of the Arctic NOx sources (e.g., oil and gas facilities, shipping) in local production and titration of O3 and in the atmospheric cycling of bromine through reactions with Br and BrO, we did not explore the regional impact of NOx emissions from local combustion sources on the Arctic surface O3 seasonal cycles systematically. This is an important aspect to further investigate particularly in light of the anticipated increase in the resource exploration in the Arctic. We have added the following (**in bold**) at the end of the second last paragraph of the conclusion section:

"… While chemistry generally leads to an overall O_3 loss in the Arctic, net production of O_3 is found to occur locally in ship plumes, downwind of oil and gas facilities in the Arctic, and in northern boreal wildfire plumes. **Interestingly, recent studies have highlighted the important role of anthropogenic NOx emissions from existing Arctic oil and gas infrastructures in perturbing O_3 and bromine chemistry, influencing the Arctic surface O_3 seasonal cycles at local and regional scales (Peterson et al., 2025; Widmaier et al., 2025). Although results from the present study do reflect the individual effects of NOx emissions from local anthropogenic sources in both production and titration of O_3 as well as in atmospheric cycling of bromine through reactions with Br and BrO, we did not explore the role of NOx emissions from local combustion sources in the Arctic surface O_3 seasonal cycles systematically. This is an important aspect to further investigate, particularly in light of the anticipated increase in the resource exploration in the Arctic under warming climate.**"

Additional comments:

L99-100: Another pertinent paper to cite is Peterson et al. 2017 (ACP, https://doi.org/10.5194/acp-17-7567-2017).

R: Thanks for the suggestion. We have added the reference to Peterson et al. (2017) here.

L109 & L568: An earlier important paper to also cite is Raso et al. 2017 (PNAS, https://www.pnas.org/doi/10.1073/pnas.1702803114).

R: Thanks for the suggestion. We have added the reference to Raso et al. (2017) here along with the reference to (Benavent et al., 2022).

L323 & Appendix 1 table: Add Jeong et al. 2022 (ACS Earth Space Chem, *https://doi.org/10.1021/acsearthspacechem.2c00189), in addition to Burd et al.*

R: Thanks for suggesting the additional reference here. Indeed, the study of Jeong et al. (2022) reached the same finding as Burd et al. (2017) regarding the termination of snowpack bromine release following spring snowmelt. We have added the reference as suggested.

Table 1: I encourage writing "(Utqiagvik)" after "BARC" (under MAX-DOAS section) for improved clarity.

R: Done.

Pages 20-21: There is a very long paragraph that extends ~1.5 pages. I encourage breaking it up to make it easier to read.

R: Thanks for the suggestion. We have reorganised the discussion in this section to make it more readable. We have moved the original discussion on the impact of the model parameterization of dry deposition on model performance at the two northern European boreal sites to combine with the discussion on statistical evaluation, as the low bias of modelled O3 is characterised there. We have also eliminated the discussion on uncertainty in the model representation of biogenic VOC emissions and its possible impact on model performance, for the sake of shortening the overall length of the paper (which was one of the general comments from Reviewer 1). The first two paragraphs of section 3.2 (L509 – 577) in the previous version of the manuscript have been reorganised into six paragraphs as follows:

"To evaluate the models' ability to simulate Arctic boundary layer O_3, the modelled surface (or lowest model level) O_3 concentrations are compared with observations from ground-based monitoring sites and surface mobile platforms (O-Buoys and Mirai cruise). To do this, the modelled O_3 concentrations are extracted at the ground-based sites and following buoy tracks and ship paths from the nearest model grid cells and hours and compared with hourly observations. Existing model evaluations related to tropospheric ozone assessment (e.g., Monks et al., 2015; Whaley et al., 2023; Young et al., 2018) have been mostly performed on long-term annual and monthly averages. With the two regional models used in this study run at much higher spatial resolutions, as compared to the global models employed in the previous assessment studies, we can examine model simulations and compare with observations at much finer temporal resolutions (e.g., hourly) here.

Figure 4 shows the O_3 time series comparisons at the eight Arctic monitoring sites described in 2.2.1. Overall, both DEHM and GEM-MACH simulations captured the observed O_3 seasonal as well as synoptic-scale variations at these Arctic ground sites. The three Arctic coastal sites, Utqiagvik,

Villum, and Tiksi, are strongly influenced by the spring ODEs, which are captured reasonably well by the GEM-MACH simulation. The DEHM model was less successful in capturing the springtime ODEs at these sites. The modelling of ODEs will be examined in more details later in section 4.1. The seasonal variation in the observed O_3 at the subarctic inland sites (Tustervatn, Pallas, and Esrange) follows the typical pattern of a maximum in spring and minimum in summer, with greater variability in summer and fall. The model simulations from both DEHM and GEM-MACH follow closely the observed O_3 variations throughout the year. The GEM-MACH simulation has a larger low bias at the two northern European boreal sites (Pallas and Esrange) particularly during the spring and summer seasons, while the DEHM performed better (particularly at Esrange); this will be discussed further in the statistical evaluation below.

The two high-elevation sites (Zeppelin and Summit) exhibit somewhat different O_3 seasonal patterns. The Zeppelin site, situated at 474 m above the Arctic Ocean, is situated approximately half of the time above the top of the atmospheric boundary layer (Dekhtyareva et al., 2018). The observed O_3 time series in 2015 displays an overall maximum in April and a minimum in July, in contrast to the Arctic coastal sites. This is consistent with the seasonal patterns based on a longer time (multi-year) observations (e.g., Whaley et al., 2023). However, it is evident from the time series in Fig. 4 that the site is sporadically impacted by springtime ODEs during April and May in 2015. Previous observations of ODEs at this site have been reported by others (e.g., Berg et al., 2003; Eneroth et al., 2007; Lehrer et al., 1997; Solberg et al., 1996). The O_3 observation at Summit has a gap between the end of July and the end of October in 2015. The incomplete observed O_3 time series shows no clear trend over the first 5 months (January – May) of 2015 before increasing over June to reach a maximum in July. This is a departure from the seasonal trend shown in Whaley et al. (2023) based on multi-year data (2003 – 2018), which showed a maximum in May. Both Zeppelin and Summit surface observations show high O_3 events in July 2015. As will be discussed later in 4.2, there is an indication that these events may be associated with transport of wildfire plumes in the free troposphere. Again, model simulations from both DEHM and GEM-MACH compare well with the observations at these sites, capturing the observed seasonal and synoptic scale variations (also evident from the statistical evaluation shown in Table 2), though neither of the model simulations was able to fully capture the July high O_3 events observed at Summit.

Statistical evaluations of model performance were conducted on the hourly time series. Table 2 shows selected seasonal and annual model performance scores at the 8 Arctic ground sites, including normalised mean bias (NMB), Pearson correlation coefficient (r), and unbiased root-mean-square-error (URMSE), while the corresponding monthly scores are shown in SF.2. The seasonal scatter plots (colour coded for each month separately) of model vs. observations at the 8 surface sites are shown in SF.3. The evaluation (Table 2) shows that both models underpredict wintertime Arctic surface ozone at all sites, with GEM-MACH having a greater negative bias at Utqiagvik, Villum, Pallas and Esrange. At coastal sites, the DEHM model has significant positive bias during the spring months due to its under-representation of the springtime ODEs, while the GEM-MACH model has considerably better performance scores. It is interesting to note the significant positive bias in both models during the summer months at the coastal sites, except for a small negative bias in GEM-MACH at Villum, which is largely driven by the month of June values; see SF.3(b). Neither DEHM nor GEM-MACH currently includes iodine chemistry, which can play a prominent role in ozone destruction over polar oceans during (as well as after) the time of

springtime bromine explosions (Benavent et al., 2022; Fernandez et al., 2024; Mahajan et al., 2010; Raso et al., 2017; Wittrock et al., 2000).

At the two northern European boreal sites, Pallas and Esrange, the models are generally biased low throughout the year. GEM-MACH has the greatest difficulty in simulating surface ozone accurately at these two sites particularly during summer as evident by the relatively poor performance scores shown in Table 2 (and SF.3) compared to other sites, while DEHM performed considerably better at these sites. This may be partly attributable to the difference in modelled O_3 dry deposition velocities over the boreal landcover between GEM-MACH and DEHM. Clifton et al. (2023) examined O_3 dry deposition velocity formulations across contemporary regional chemical transport models, including the formulations used in GEM-MACH (based on Wesely, 1989) and DEHM (as in Simpson et al., 2012). They showed that the formulation used in GEM-MACH ("GEM-MACH Wesely") significantly overestimated O_3 dry deposition velocities over the European boreal forest during summer compared to estimates based on ozone flux measurements. In contrast, the formulation used in DEHM ("DO3SE") was shown to produce O_3 dry deposition velocities in much closer agreement with those derived from observations over the European boreal forest in summertime.

Overall, the two regional models seem to demonstrate better skill in capturing the observed seasonal variations in the Arctic surface ozone, compared to the large-scale global atmospheric chemistry models reported in previous assessments (e.g., Law et al., 2023; Whaley et al., 2023; Young et al., 2018) where the models showed a large spread in simulated surface O_3 concentrations and inability to reproduce the observed seasonal cycles at some of the Arctic sites. Besides the implementation of the processes involved in springtime ODEs in the Arctic, the better performance from the two independent regional models in this study can be attributed, at least in part, to better resolved atmospheric dynamics and boundary layer processes modelled at finer spatial and temporal scales."

Figure 4: While the colors are described in the caption, I encourage adding a legend as well for the three lines.

R: Thanks. Done.

Figure 5: This figure was confusing at first, as I initially couldn't figure out why the gray traces were different between the plots. To improve clarity, I suggesting removing the header "O-Buoy & Mirai" and replacing with "Observations", "GEM-MACH", and "DEHM" above each plot, to emphasize that this is the comparison being shown.

R: Thanks for the suggestion. We have fixed the plots as suggested.

Figure 6: It would be helpful if the gray background behind the entirety of the plots could be removed. It would also be helpful if font sizes could be increased. Both of these edits should improve readability. Define the month abbreviations in the caption.

R: We have improved the figure quality, and the clarity of the caption as suggested.

L694: Moore et al. 2014 (Nature, https://doi.org/10.1038/nature12924) is an important paper to cite here for convection-based springtime ozone recovery.

R: Thanks for suggesting this – a very relevant paper to refer to here. We have added the reference to Moore et al. (2014) here.

L696-699: This speculation can be supported by prior observations of reactive bromine in Utqiagvik in February by Custard et al. 2017 (ACS Earth Space, https://doi.org/10.1021/acsearthspacechem.7b00014) and Simpson et al. 2018 (GRL, https://doi.org/10.1029/2018GL079444). For context, polar sunrise occurs at Utqiagvik in late January.

R: Thanks for the suggestion. We have added the following sentence:

"The release of reactive bromine from snowpacks at this location during early spring is supported by observations (e.g., Custard et al., 2017; Simpson et al., 2018)."

L715: It would seem that the improved simulation of the ODEs only at the Zeppelin site is associated with the site being above the inversion layer, as opposed to the other sites near sea level. This seems worth noting.

R: The discussion here is directed at the DEHM simulations (considering blowing-snow sourced bromine only) being less successful in capturing the observed ODEs (except for a few occasions in late April when DEHM was able to capture the observed ODEs at the Zeppelin site). It is not clear whether this is attributable to the Zeppelin site being located above the inversion layer as opposed to the other coastal sites close to the sea level.

Figure 8: It would be helpful to increase the font sizes, especially the y axis labels, to make them more readable. Also, the wind vector symbols are very small and difficult to discern.

R: Thanks. We have improved the figure quality (legibility).

L753: I suggest citing Oltmans et al 2012 (JGR, https://doi.org/10.1029/2011JD016889) and Seabrook et al 2011 (JGR, https://doi.org/10.1029/2011JD016335) as excellent examples of vertical ozone profiles over extended field campaign periods.

R: Thanks for suggesting these two papers here. They both provide observations on the vertical structure of ODEs and should be referenced here. We have added the following sentences:

"Using a differential absorption LIDAR, Seabrook et al. (2011) observed the vertical structure of springtime ODEs over the Arctic Ocean off the south coast of Banks Island. They found that the observed ODEs were largely confined within the lowest 200-600 m of the atmosphere and were associated with airmasses being in contact with sea ice for an extended period. Oltmans et al. (2012) analysed the vertical profiles from the near-daily ozonesonde measurements conducted during 2008 and 2009 spring periods at Barrow (Utqiaġvik) and found that the depletion was confined to approximately the lowest 1000 m with an average height of the top of the layer at ~500 m."

Figure 9: Please increase the date font size in the center map figure, as well as the outer legend font size.

R: Done.

Figures 12, 15, 16, 18: The upper left label, above the colorscale, on each plot is not readable. Please reformat to increase font size, or remove if the label is the same as the top header on the figure. For Figure 18, the larger font (right corner labels) currently do not include units.

R: We have improved the legibility of the labelling on these plots and provided the unit in figure caption for Figure 8.

Figure 17: It would be helpful to increase linewidths of the font throughout (or make them bold) to make the text easier to read.

R: Done.

L1105: Simpson et al 2017 (ACP, https://doi.org/10.5194/acp-17-9291-2017) is an important paper to cite here, as it points to the importance of aerosols and needed chemical composition measurements, since the presence of aerosols alone did not the reactive bromine.

R: Thanks for the suggestion. However, we did not explore the uptake dependency on aerosol composition in heterogeneous cycling of reactive bromine. This may be pursued in our future studies. We have added a sentence here:

"Simpson et al. (2017) also found that higher aerosol extinction (> 0.1 km^{-1}) appeared to be necessary for maintaining the notable presence of BrO aloft, though they suggest that chemical composition of aerosols may play a role as well in the cycling of reactive bromine."

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