Authors' response to reviewers' comments

We thank both reviewers for their helpful and constructive comments. Our responses to the reviewer's comments are included below following each reviewers' comment. The reviewers' original comments are shown in *italic*, our responses are in blue, and the corresponding changes are specified in red. We have added designators/numbering to the reviewers' comments for cross referencing (in the Annex – List of changes attached at the end).

Reviewer 1 (R1)

This manuscript describes a detailed evaluation of Arctic tropospheric ozone in two regional chemical transport models using a variety of surface and vertical profile measurements for the year 2015. The study undertakes a thorough observational comparison, followed by a detailed investigation of the role of halogen chemistry in controlling model ozone, an analysis of wildfire emission contributions, and finally a regional Arctic tropospheric ozone budget analysis from one of the two models.

Among other aspects, the detailed comparison of high resolution ozone simulations with hourly data is an important advance on many previous studies that have evaluated coarse global-scale models with monthly mean observations. The presentation of comparison of model simulations that include detailed halogen chemistry against simulations with this removed is also very informative, as are the investigations of sensitivity to assumptions in the model Br mechanisms. In addition, evaluation of the structure of modelled and observed ozone vertical profiles using aircraft observations during springtime is of great benefit. These results are of high value to the Arctic atmospheric composition and modelling communities.

Generally, I do not have any major concerns or reservations with the paper. The analysis presented is very thorough, and the manuscript is well written. I recommend that the manuscript is suitable for publication in ACP once the following comments have been addressed.

General comments (GC)

R1-GC1. The paper is very long, however I recognise that the analysis presented is very thorough. I have made one suggestion of where text could be shortened by avoiding separation of parts of the results that could be better linked (see below).

R: See response to the specific comments below.

R1-GC2. In a couple of places more could be done to compare the performance of the models presented here with previous model assessments of ozone using the same datasets. I have highlighted a couple of examples below in my specific comments.

R: See responses to the specific comments below.

Specific comments (SC)

R1-SC1. Line 61: Archibald et al., (2020) is not a primary reference for the impacts of ozone on health and ecosystems. Can the authors provide alternative references for these two aspects of ozone impact.?

R: We have updated the references to those pertinent to the impact of ozone on health (Fleming et al., 2018; US Environmental Protection Agency, 2013; World Health organization, 2013) and ecosystems (Ainsworth et al., 2012; Mills et al., 2011, 2018).

Change (revised text):

"Tropospheric ozone (O_3) is a greenhouse gas (GHG) and, near the surface, an air pollutant harmful for human health (Fleming et al., 2018; US Environmental Protection Agency, 2013; World Health organization, 2013) as well as affecting crop and ecosystem productivity (Ainsworth et al., 2012; Mills et al., 2011, 2018)."

R1-SC2. Line 65: "changes [..].. in the transport pattern from lower latitudes" This needs to be more explicit to provide context. i.e. "changes in the patterns of transport of ozone and precursors from lower latitudes".

R: We have incorporated the reviewer's suggestion.

Change (revised text):

"... as a result, changes in local anthropogenic and natural sources of O_3 precursors and in the patterns of transport of O_3 and its precursors from lower latitudes as well as increased vertical mixing are to be expected."

R1-SC3. Line 88: "The ability of current models to simulate Arctic tropospheric O3 has been evaluated in several studies (e.g., Monks et al., 2015b; Shindell et al., 2008; Whaley et al., 2023)" What is meant by "current" in this context (given that citations from 2015 are relevant here)? I agree with the need to cite some of these older studies, since it is not clear that the models have improved substantially in this time. Maybe omit "current" and rephrase as "... evaluated in several previous and recent studies..."

R: We have rephrased as suggested by the reviewer.

Change (revised text):

"The ability of models to simulate Arctic tropospheric O_3 has been evaluated in several previous and recent studies (e.g., Monks et al., 2015b; Shindell et al., 2008; Whaley et al., 2023) involving largely global models."

R1-SC4. Line 190-193: Does this imply that within the European domain ECLIPSE emissions are not used (replaced by EMEP)? What is the motivation for this? Is it simply more information from higher resolution? How different are the emissions?

R: It is correct that over Europe the ECLIPSE emissions were replaced by the EMEP emissions in the DEHM simulations. There were several reasons for this replacement. Over Europe, the EMEP emission inventories have higher spatial (0.1° x 0.1° as compared to ECLIPSE at 0.5° x 0.5°) and temporal resolution (EMEP provides yearly emissions from 1990 to 2022 as compared to the 5 years intervals from 1990 to 2020 for ECLIPSE). ECLIPSE6b was developed in 2018-2019, while the EMEP emissions are continuously updated each year not only for the newly added year but also for previous years retrospectively (i.e., previous inventories are revised when new activity data for the different European countries or updated emissions factors are available). There are differences in emissions between the ECLIPSE and EMEP inventories over Europe both in spatial distribution (due to the difference in spatial resolution and allocation) and in total amount (most pronounced for NOx and NMVOC, where ECLIPSE emissions are significantly higher than EMEP emissions). The DEHM model and other European models such as the EMEP model have intensively used and tested the EMEP emissions in many projects, where the model simulations for Europe shows good performance (Brandt et al., 2012; Simpson et al., 2012). We have revised the text to "The anthropogenic emissions from the ECLIPSE v6b dataset at 0.5° x 0.5° resolution (Klimont et al., 2017) are used for the portion of the model domain outside Europe, while for the areas over Europe the emissions from the European Monitoring and Evaluation Programme (EMEP) expert database with 0.1° x 0.1° resolution are used (see https://www.ceip.at/)".

Change (revised text):

"The anthropogenic emissions from the ECLIPSE v6b dataset at 0.5° x 0.5° resolution (Klimont et al., 2017) are used for the portion of the model domain outside Europe, while for the areas over Europe the emissions from the European Monitoring and Evaluation Programme (EMEP) expert database with 0.1° x 0.1° resolution are used (see https://www.ceip.at/)."

R1-SC5. Table 1: It would be useful to add to this table the temporal resolution of the data measured and/or used in the study. Could this perhaps be added into column 3? Similarly for sondes, what is the approximate vertical resolution of the data?

R: We have added the appropriate information on data temporal resolution in Table 1. For ozonesondes, the data obtained from HEGIFTOM has the vertical resolution varying from a few meters to a few tens of meters over the lowest 5 km of the atmosphere. We have added this information at the end of section 2.2.3.

Changes:

- 1. We have added the observational data frequency information in Table 1 (3rd column) for ground sites, buoys, ship, and MAX-DOAS (BrO) data.
- We added the following sentence at the end of section 2.2.3:
 "The vertical resolution of the ozonesonde data varies between a few meters to a few tens of meters (< 50 m) over the lowest 5 km of the atmosphere."

R1-SC6. Section 3.1 and Fig. 2 discussion. There is no mention of the low ozone simulated in both models over the northern Eurasian region during winter. This is also evident in Figure 3, which highlights the winter months as being the time of the minimum at the surface. Is this the impact of ozone titration by Eurasian NO emissions in winter?

R: We have added "Both models simulated low surface O3 concentrations over northern Eurasia and northern Europe during winter. The low ozone can be argued to be attributable to reduced photo-chemical production and enhanced titration by NO emissions from local sources within the darker and shallower boundary layer during winter, as well as dry deposition". This is supported by examining the O3 tendencies from one of the models (GEM-MACH) as discussed in section 4.3.

Change (added text):

"Both models simulated low surface O₃ concentrations over northern Eurasia and northern Europe during winter. The low ozone can be argued to be attributable to reduced photo-chemical production and enhanced titration by NO emissions from local sources within the darker and shallower boundary layer during winter, as well as dry deposition."

R1-SC7. Figure 4 - Would it be possible to add a legend to the figure labelling the coloured lines used?

R: We have added the legend in the figure.

Change: the revised Figure 4 now include legend for the coloured lines.

R1-SC8. Line 557: This text describes the statistical evaluations for the comparisons shown in Fig. 4. I am not sure this needs to be separated from the presentation of performance of the models in the previous paragraph. The text could be combined to reference the statistics as part of the discussion of model performance. This would also help qualify several subjective terms such as "compare well" (e.g. line 551).

R: Here we present the comparison of O3 time series first to examine how model simulations are capable of capturing the seasonal and synoptic variabilities in surface O3 observed at the Arctic sites in a broad sense. This is followed by statistical evaluation to quantify and characterise model performance, which leads to discussions on several processes affecting the model performance. We tried to combine the statistical evaluation with the discussion on time series comparison as suggested by the reviewer, but it did not improve the clarity and readability. However, we have reorganised this section, moving 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 model representation of biogenic VOC emissions and its possible impact on model performance, for the sake of shortening the length of the paper. This reorganisation was also in response to Reviewer 2's suggestion to break up the long opening paragraph of 3.2. Regarding the subjective term "compare well" used in describing the time series comparison, we have added more explicit descriptives such as, "capturing the observed seasonal and synoptic scale variation (also evident from the statistical evaluation shown in Table 2)".

Changes:

- 1. Reorganised section 3.2, splitting the previous two long paragraphs into 6 paragraphs, moved the 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, removed the discussion on uncertainty in model representation of biogenic VOC emissions and its possible impact on model performance, for the sake of shortening the length of the paper. See response to Reviewer 2's comment (R2-AC5) below.
- 2. Revised text to be specific about what we mean by "compare ell":
 "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 model
 simulations was able to fully capture the July high O₃ events observed at Summit."

R1-SC9. Line 575: The authors make the statement that the comparisons shown demonstrate improved model performance compared with similar evaluations using global models. Would it be possible to be more quantitative, given that previous studies have used the same surface sites for evaluation and will have quoted e.g. mean bias values (notwithstanding the use of different time resolution data)?

R: The statement was made largely based on a qualitative comparison between similar model-observation time series from the present study and those presented in previous studies mostly comprised of large-scale global models (e.g., Monks et al., 2015; Whaley et al., 2023; Young et al., 2018). Unfortunately, very little standardized statistics for these models have been presented in such a way that can be compared quantitatively with the results in this study. Only Whaley et al. (2023) provided annual (normalised) mean bias from the multi-model ensemble at selected Arctic ground sites, based on monthly mean values (see their Figure 6). Such a metric has issues with error cancellations amongst models and seasonal variations which can give a false impression of accuracy. Ideally, a suite of robust metrics which can more accurately identify the true seasonal and sub-seasonal variations and associated errors between models and observations is desired. We have revised the text to "Overall, the two regional models seem to demonstrate better skills 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 large spread in simulated

surface O3 concentrations and inability to reproduce the observed seasonal cycles at some of the Arctic sites. Besides the implementation of the processes involved in springtime bromine chemistry 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".

Change (revised text):

"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."

R1-SC10. Section 3.1: The Whaley et al., (2023) study presented evaluation of a set of global models against ozone sonde data (Figure 8 in their paper). It would be informative to make some sort of reference / comparison to this in putting the results presented by the authors into context.

R: We believe that the reviewer is referring to section 3.3 (Ozone vertical profiles comparison with ozonesondes). Yes, a reference to their study would be informative on how the regional models in this study compare with the large-scale models in that study. To do this, we have included a new figure in supplementary material (SF. 5; in the revised version) showing similar plots of relative difference (or the normalised mean bias, NMB) between modelled and observed ozone profiles which can be directly compared to their study. Again, the simulated O3 from the two regional models included in this study are shown in much better agreement with the ozonesonde observations than the large-scale models in Whaley et al. (2023), with significantly smaller relative difference (or NMB), generally well within +/-25% over the lowest 5 km of the atmosphere, compared to +/-50% shown in Whaley et al. (2023). We have added a short paragraph on this in section 3.3 of the revised manuscript.

Changes:

- 1. Added a plot of profiles of seasonal relative difference between model simulations and ozonesonde observations in supplementary materials (SF.5), which can be compared with Whaley et al. (2023).
- 2. Added the following paragraph in 3.3:

 "Whaley et al. (2023) compared model simulated vertical profiles (using monthly mean model output) from 12 different large-scale models to the ozonesonde measurements at the same group of sites as we examined here (see their Figure 8 and Figure S1). We have plotted the profiles of seasonal relative difference between model simulations and observations (or NMB) in SF.5, which can be compared with the results shown in Whaley et

al. (2023). Again, the two regional models here show better skills in simulating the observed O3 vertical profiles over the lowest 5 km of the atmosphere examined (having considerably smaller biases, generally well within +/-25%, compared to the large spread of relative difference, +/-50%, in the same altitude range amongst the large-scale global models)".

R1-SC11. Figure 10 - A minor point, but maybe it is worth spelling out "interquartile range" (IQR) in the legend or caption.

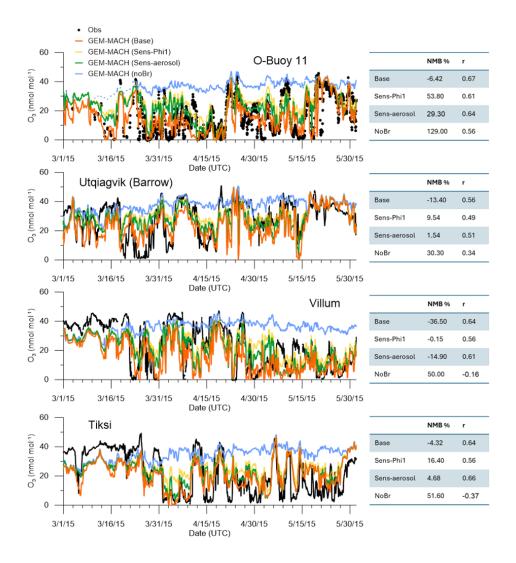
R: We have spelled out the inter-quartile-range in the caption in the revised version.

Change: added "boxes are inter-quartile-range (IQR)" in Figure 10 caption.

R1-SC12. Figure 11 - It might help in comparison of the different sensitivity simulations to provide some quantitative metrics for the comparisons with observations (i.e. mean bias / r2 values).

R: We have added a couple of statistical metrics (normalised mean bias and Pearson correlation coefficient) for reference (in revised Figure 11). However, Figure 11 is mainly to illustrate how the model's ability in simulating the observed ODEs is influenced by parameter settings that controls the snowpack bromine production (Φ_1) and bromine production through heterogeneous reactions on atmospheric aerosols (aerosol surface area). A more pertinent approach would be to design metrics to quantify how the frequency and intensity of the observed ODEs being replicated by model simulations. This will be pursued in our future investigations.

Change: revised Figure 11 now containing normalised mean bias and Pearson correlation coefficient.



R1-SC13. Line 955 - The Arnold et al., (2015) evaluation of fire-impacted O3/CO enhancement ratios are also based on monthly mean large-scale Arctic enhancements, so these could be more directly compared with results presented here (i.e. they are also not plume specific enhancements).

R: Thanks for pointing this out. It is true that Arnold et al. (2015) evaluation of the O3-to-CO enhancement ratios due to boreal wildfires were based on modelled monthly mean. However, their Δ O3/ Δ CO was evaluated from a linear fit to the scatterplot of O3-to-CO over model grids identified as dominated by wildfire influences (determined by means of fixed-lifetime CO tracers introduced in their model simulations to track transport of tracers emitted from wildfire TR_{fire} and from anthropogenic sources $TR_{anthrop}$; fire-impacted grids are those where $TR_{fire}/(TR_{fire}+TR_{anthrop}) > 0.66$). In our case, the enhancement ratios Δ O3/ Δ CO were directly calculated from the pair of model runs with and without the wildfire emissions, similar to the approach of Pfister et al. (2006) and Thomas et al. (2013). Pfister et al. (2006) also examined different approaches in evaluating the enhancement ratios, namely, an approach using a linear fit to the O3-CO scatterplot (similar to Arnold et al., 2015) vs. an enhancement approach based on excess mixing ratios of O3 and CO

(similar to this study). They showed that when the variability in the background concentration levels was well characterised, the enhancement approach would be more robust and accurate in evaluating the fire-influenced $\Delta O3/\Delta CO$ ratios than the approach based on scatterplot. The latter is subjected to the uncertainty in isolating the enhancement due to wildfires from those due to non-wildfire (anthropogenic) emissions, as a result of different NOx/CO emissions ratios between boreal wildfires and anthropogenic sources. We have replaced the previous sentence, "Note, however, here the excess ratios are evaluated based on monthly mean over a broad area while the previously reported values were mostly evaluated within plumes and for a short time period (e.g., duration of field campaign)" with a brief discussion on the large variability in reported O3-to-CO enhancement ratios attributing to boreal wildfires and the impact of different methodologies in estimating the enhancement ratios.

Change (revised text): replacing the last sentence of the first paragraph (previous version) of 4.2 with the following:

"The large variability in estimated wildfire impacted $\Delta O_3/\Delta CO$ enhancement ratios from various studies can arise from the different approaches used in evaluating the enhancement ratios. By comparing between a scatter technique (based on a linear fit to the O_3 -CO concentration scatterplot) and an enhancement technique (based on the evaluation of O_3 and CO excess mixing ratios due to wildfire emissions), Pfister et al. (2006) showed that the $\Delta O_3/\Delta CO$ ratios evaluated using the scatter technique were affected by the selection of biomass-burning-impacted air masses and the degree of mixing in the considered air masses. Much higher enhancement ratios were found in anthropogenic-combustion-impacted air masses than in the boreal-wildfire-impacted air masses, due to the difference in NOx/CO emissions ratios between these source types. Pfister et al. (2006) also showed that when the variability in the background concentration levels was well characterised (which is not a trivial task for the analysis of observational data while being quite straightforward for the analysis of model results through sensitivity runs like ours), the enhancement technique would be more robust and accurate in evaluating the fire-influenced $\Delta O_3/\Delta CO$ enhancement ratios."

R1-SC14. Page 47: Discussion of PAN/CO enhancement ratios. In the Arnold et al., (2015) study, a difference in PAN/CO enhancement values was identified between models forced using different reanalyses products (models forced using GEOS-5 data displayed lower enhancements compared with models forced by ERA-Interim data). It would be interesting to know how the models presented compare and if they are consistent with the Arnold et al., (2015) values according to the meteorological dataset used (for DEHM using ERA-5 for example).

R: The DEHM meteorology is generated by WRF driven by ERA5 reanalysis. However, as the sensitivity simulation on wildfire emissions was not performed by the DEHM group, we were not able to carry out the same wildfire impact analysis on the DEHM simulations. The Δ PAN/ Δ CO enhancement ratios due to boreal wildfires from this study (based on GEM-MACH simulations), ~3 –4 pptv/ppbv at the lowest model level, increasing with height to 6 – 7 pptv/ppbv near 700 hPa, are comparable to the PAN/CO enhancement ratios reported in Arnold et al. (2015) from the group of models driven by the ECMWF meteorological reanalysis. We have added this reference in the revised manuscript.

Change (revised text in discussion of Figure 16, section 4.2):

"... The modelled PAN enhancement ratios (Δ PAN/ Δ CO) due to boreal wildfires are simulated to be ~3 – 4 pptv/ppbv over the North American boreal fire regions at the lowest model level, increasing with height to 6 – 7 pptv/ppbv near 700 hPa, comparable with the Δ PAN/ Δ CO ratios reported by Arnold et al. (2015) from the group of models driven by the ECMWF meteorological reanalysis. ..."

Editorial / typographical corrections (EC)

R1-EC1. Line 79: "variations in the Arctic tropospheric O3" Omit "the".

R: revised as suggested.

R1-EC2. Line 458: Better as "...varying degrees of complexity."

R: revised as suggested.

Reviewer 2 (R2)

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.

Specific comments (SC)

R2-SC1. 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 a brief description of the blowing snow parameterization implemented in DEHM in the revised manuscript.

Change (revised text in 2.1.3, representation of bromine sources in DEHM):

"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 formulations 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). Similar to the implementation in Yang et al. (2010), 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 change the age of the surface snow. ..."

R2-SC2. 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 a few sentences addressing this aspect at the end of the second last paragraph of the conclusion section.

Change (added text 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 (AC):

R2-AC1. 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.

Change: added Peterson et al. (2017) to the references for bromine cycled through heterogeneous chemistry on aerosol surfaces (middle of the third paragraph in the introduction section).

R2-AC2. 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).

Change: added the reference to Raso et al. (2017) along with the reference to Benavent et al. (2022) in the last sentence of the third paragraph of the introduction section.

R2-AC3. 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.

Change: added the reference to Jeong et al. (2022) along with the reference to Burd et al. (2017) related to the termination of snowpack bromine release following snowmelt (2nd paragraph under the heading "representation of bromine source in GEM-MACH" in 2.1.3, and in Appendix 1).

R2-AC4. Table 1: I encourage writing "(Utqiagvik)" after "BARC" (under MAX-DOAS section) for improved clarity.

R: Done.

Change: added Utqiagvik after BARC in Table 1, as well as in 2.2.5.

R2-AC5. 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.

Change: revised/reorganised text in section 3.2, first 6 paragraphs in the revised manuscript (replacing the previous first two paragraphs of 3.2):

"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, Utqiagʻvik, 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 detail 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 shows 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 model evaluation below.

The two high-elevation sites (Zeppelin and Summit) exhibit somewhat different O₃ 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 O₃ 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 O₃ observation at Summit has a gap between the end of July and the end of October in 2015. The incomplete observed O₃ 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 display high O₃ 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 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 rootmean-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 versus 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 an estimate 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 summertime European boreal forest.

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."

R2-AC6. Figure 4: While the colors are described in the caption, I encourage adding a legend as well for the three lines.

R: Thanks. Done.

Change: added legend for the coloured lines in the revised Figure 4.

R2-AC7. 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.

Change: revised the headings for the plots in Figure 5.

R2-AC8. 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.

Change: revised plots in Figure 6 to improve quality and added definition for the month abbreviations in the figure caption.

R2-AC9. 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.

Change: added the reference to Moore et al. (2014) along with other references related to O3 concentration recovery via vertical and horizontal air mass exchanges (2nd paragraph of 4.1).

R2-AC10. 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)."

Change: added the above sentence in the 2nd paragraph of 4.1:

"... There were a few brief episodes of depletion in early March observed at Utqiagʻvik when surface O_3 concentrations decreased by about 20 ppbv from the background level of 30 - 40 ppbv to about 10 ppbv, which may well be associated with bromine chemistry given its relatively southern location (71.32°N, hence having more than 10 hours daylight by early March) (Frieß et al., 2011) and its proximity to FY sea ice. 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). ..."

R2-AC11. 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.

R2-AC12. 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).

Change: revised Figure 8 to improve clarity of the plots (including increased font size of the axis labels and the legibility of the vectors).

R2-AC13. 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 of time. Oltmans et al. (2012) analysed the vertical profiles from the near-daily ozonesonde measurements conducted during 2008 and 2009 spring periods at Barrow (Utqiagʻvik) and found that the depletion was confined to approximately the lowest 1000 m with an average height of the top of the layer at \sim 500 m."

Change: The above sentences have been incorporated into the 3rd paragraph in 4.1:

"While there are relatively abundant surface observations of the Arctic springtime ODEs from the ground-based monitoring sites and mobile platforms (e.g., buoys and research vessels) in the Arctic Ocean (Bottenheim et al., 2009), observations on the vertical structure of ODEs are relatively scarce. 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 of time. Oltmans et al. (2012) analysed the vertical profiles from the near-daily ozonesonde measurements conducted during 2008 and 2009 spring periods at Barrow (Utqiagvik) 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."

R2-AC14. Figure 9: Please increase the date font size in the center map figure, as well as the outer legend font size.

R: Done.

Change: revised Figure 9 to improve clarity of the plots, including increased font size of date label (centre map), axis labels, and legend.

R2-AC15. 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 18.

Changes:

Figure 12 – improved labels on individual plots (above the colour scale) and clarified units in the labels above each column and in the figure caption.

Figure 15 – improved labels on individual plots (above the colour scale) and specified units in the labels above each column.

Figure 16 – removed the illegible labels above the colour scale in the individual plots and added units to the variable labels at the top of each column.

Figure 18 – improved labels on the individual plots (above the colour scale) and provided the units for the tendency terms (AO3, GO3, UO3, and smO3) in the figure caption.

R2-AC16. 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.

Change: revised Figure 17 to improve legibility, including increased line thickness and increased font size of the axis labels.

R2-AC17. 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."

Change: revised text towards the end of the 4th paragraph of section 5 (conclusion) to include the above sentence:

"... The study also demonstrates that atmospheric aerosols play an integral role in the Arctic springtime bromine explosions and ODEs through heterogeneous cycling of reactive bromine, particularly over a deeper vertical layer and at distance from the snowpack bromine source area. 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. This has implications for the potential role of Arctic haze aerosols that may play in the springtime ODEs, as indicated in previous studies (e.g., Fan and Jacob, 1992)."

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Annex: List of changes

Abstract

Minor editorial wording changes

1 Introduction

- Updated references in the first paragraph (Fleming et al., 2018; US Environmental Protection Agency, 2013; World Health organization, 2013) and (Ainsworth et al., 2012; Mills et al., 2018), in response to reviewer 1's comment (R1-SC1)
- Added references Peterson et al. (2017) and Raso et al. (2017) in paragraph 2 in response to comments from reviewer 2 (R2-AC1 and R2-AC2)
- Minor wording changes in response to comments from Reviewer 1 (R1-SC2, R1-SC3, and R1-EC1)
- Other editorial/typo corrections

2 Study method

2.1.1 DEHM

Revised text in paragraph 3 in response to reviewer 1's comment (R1-SC4)

2.1.3 Model representations of bromine source mechanisms in the Arctic

- Minor wording change "transformation" to "oxidation" in 1st paragraph

Representation of bromine source in GEM-MACH

- Added reference to Jeong et al. (2022) in paragraph 2 in response to reviewer 2's comment (R2-AC3)
- Wording change in the last sentence for clarification

Representation of bromine sources in DEHM

 Added a brief description of the blowing snow parameterization implemented in DEHM in the second paragraph, in response to reviewer 2's comment (R2-SC1)

2.2 Observations used in this study

 Table 1: added observational data frequency information in column 3, in response to reviewer 1's comment (R1-SC5); added Utqiagvik after BARC as suggested by reviewer 2 (R2-AC4)

2.2.3 Ozonesondes

- Minor editorial corrections
- Added a sentence specifying the vertical resolution of ozonesonde data, in response to reviewer 1's comment (R1-SC5)

2.2.5 MAX_DOAS BrO VCD data

Minor editorial corrections

- Added "Utqiagvik" after "BRAC" as suggested by reviewer 2 (R2-AC4)

3.1 Seasonal distribution of lower tropospheric O₃ in the Arctic

- Minor editorial changes including responding to reviewer 1's suggestion (R1-EC2)
- Added two sentences on model-simulated low surface O₃ over northern Eurasia and northern Europe during winter, in response to a comment from reviewer 1 (R1-SC6)
- Wording change in the last sentence of the 1st paragraph to improve clarity

3.2 Annual O₃ time series comparison with observations

- The first part of the section (the first two paragraphs in the previous version) was reorganised into 6 (shorter) paragraphs, including moving the discussion on the impact of model parameterization of dry deposition on model performance over the two European boreal sites from previously the discussion segment on time series comparison to the discussion segment on statistical evaluation and the removal of the discussion regarding model representation of biogenic emissions (for the sake of shortening the overall manuscript length), to jointly address comments from reviewer 2 (R2-AC5) and reviewer 1 (R1-SC8 and R1-SC9).
- Added legend for the coloured lines in Figure 4, in response to suggestions from both reviewers (R1-SC7 and R2-AC6).
- Revised the headers in Figure 5, as suggested by reviewer 2 (R2-AC7).
- Other minor editorial wording changes to improve clarity.

3.3 Ozone vertical profiles comparison with ozonesondes

- The last sentence of the 1st paragraph is modified to reflect the removal of the discussion on the potential impact of model representation of biogenic VOC emissions on model performance at the two northern European boreal sites from section 3.2 (see above).
- A new short paragraph (2nd paragraph) is added to discuss model's ability in simulating ozonesonde observations in comparison with the results from large models included in Whaley et al. (2023), along with an additional figure (SF.5), showing seasonal vertical profiles of relative difference between model and ozonesonde observations, in the supplementary material. These changes are in response to reviewer 1's comment (R1-SC10).
- Improved Figure 6, including removing the grey background and increased font size of axis labels, as well as added definition of month abbreviations in the caption of Figure 6, in response to reviewer 2's comment (R2-AC8)

4.1 Modelling springtime ODEs: sensitivities to process representations and their uncertainty

- Added a reference to Moore et al. (2014) along with other references relating to O₃ recovery due to vertical and horizontal air mass exchange in 2nd paragraph, in response to reviewer 2's comment (R2-AC9).
- Added a 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)", in the second paragraph, in response to a comment form reviewer 2 (R2-AC10).

- Added two sentences (see below) describing the modelled surface BrO concentration time series extracted from a neighbouring grid at the Utqiagvik site shown in Figure 8(a) (red dashed line), which is relevant to reviewer 2's comment (R2-SC2).

"Note that in Fig. 8(a) the GEM-MACH-simulated surface BrO from both the grid nearest to the Utqiagvik site and a neighbouring grid (red dashed line) are plotted (third row). The lower BrO simulated at the Utqiagvik grid (compared to the neighbouring grid) is due to the higher NO₂ from local sources, which depletes BrO (to form BrONO₂) efficiently."

- Two sentences are added to the beginning of the 3rd paragraph (following the first sentence) to refer to two previous studies, Seabrook et al. (2011) and Oltmans et al. (2012), featuring observations on the vertical structure of ODEs, in response to reviewer 2's comment (R2-AC13).
- Two sentences (se below) are added referencing the work of Brockway et al. (2024) showing that reactive bromine responsible for depleting ozone is found in a very shallow stable boundary layer of a few hundred metres over the Alaska North Slope region and over the Beaufort Sea snow-covered sea ice.

"Brockway et al. (2024) describe that BrO (and thus reactive bromine that depleted O_3) over the Alaska North Slope region and over the Beaufort Sea snow-covered sea ice occurred in a shallow, very stable boundary layer up to just a few hundred meters. Occasionally they observed some lofted bromine, but mostly that was below 300m."

- Figure 8: increased font size of the axis labels and improved legibility of the vector plot, in response to reviewer 2's comment (R2-AC12).
- Figure 9: increased font size of the date labels (centre map), axis labels, and legend, in response to reviewer 2's comment (R2-AC14).
- Figure 10: added spelled-out IQR, inter-quartile-range, in the figure caption, as suggested by review 1 (R1-SC11).
- Figure 11: added simple statistical model evaluation scores (normalised mean bias, NMB, and Pearson correlation coefficient) in response to reviewer1's comment (R1-SC12).
- Figure 12: improved clarity of the labels on the individual plots, in response to reviewer 2's comment (R2-AC15).

4.2 Impact of boreal wildfires on summertime Arctic O₃

- Revised text at the end of paragraph 1 in discussing the context of how the boreal-wildfire-induced O₃-to-CO enhancement ratios found in this study compared to those reported in previous literatures, in response to reviewer 1's comment (R1-SC13).
- Added a phrase in paragraph 2 on the wildfire-induced PAN-to-CO enhancement ratio from this study being comparable to those reported in Arnold et al. (2015) from the group of models driven by the ECMWF meteorological analysis, in response to reviewer 1's comment (R1-SC14).
- Figure 15: improved legibility of labels on individual plots in response to reviewer 2's suggestion (R2-AC15).

- Figure 16: improved legibility of labels on individual plots in response to reviewer 2's suggestion (R2-AC15).
- Figure 17: increased line thickness and increased font size of the axis labels to improve legibility, in response to reviewer 2's suggestion (R2-AC16).
- Minor editorial wording changes to improve clarity.

4.3 Ozone tendency and budget analysis

- Figure 18: improved legibility of labels in the individual plots and specified the units for the tendency terms in the figure caption, in response to reviewer 2's comment (R2-AC15).
- Minor editorial wording changes to improve text clarity.

5 Conclusions

- Added a sentence in the 3rd paragraph on the role of reactive bromine cycling through heterogenous reactions on aerosol surfaces (referring to Simpson et al., 2017), in response to reviewer 2's comment (R2-AC17).
- Added a sentence in paragraph 4 pointing out that the model-simulated direct photochemical production of molecular bromine from snowpacks, which is responsible for ozone depletion, is consistent with what is believed to occur from several experimental studies (Custard et al., 2017; Halfacre et al., 2019; Pratt et al., 2013).
- Added a few remarks at the end of paragraph 6 on the role of anthropogenic NOx emissions from existing Arctic oil and gas infrastructures in perturbing O₃ and bromine chemistry, influencing the Arctic surface O₃ seasonal cycles at local and regional scale, which was highlighted in recent publications (Peterson et al., 2025; Widmaier et al., 2025), and to point out that although this aspect was not explored systematically in the present study, the individual effects of anthropogenic NOx emissions were reflected in the results from our study. This is in response to the reviewer 2's specific comment (R2-SC2).

Appendix 1 Model key features and configuration

- Added reference to Jeong et al. (2022) in the describing bromine chemistry and source representation in GEM-MACH (related to the termination of bromine release from snowpacks), in response to reviewer 2's comment (R2-AC3).