

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2022-44

Anonymous Referee #2

Referee comment on "Comparison of model and ground observations finds snowpack and blowing snow aerosols both contribute to Arctic tropospheric reactive bromine" by William F. Swanson et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2022-44-RC2>, 2022

GENERAL COMMENTS

This study uses the chemical transport model GEOS-chem to assess the relative importance of two major halogen sources in the Arctic, one from snowpack and the other from sea salt aerosol generated by blowing snow. Modelled BrO (and ozone) concentrations are compared to observations at Utqiagvik/ Alaska and within the sea ice at three drifting O-Buoy platforms during spring 2015. The main findings are that the snowpack source has a larger impact on BrO than the SSA source from blowing snow, but both mechanisms are active in the Arctic and are needed to achieve the best match between model and observations.

The authors are commended for a valuable scientific contribution as this is only the second model study after Marelle et al., 2021 to include both halogen source mechanisms in one model, which allows for a meaningful assessment of relative importance of source mechanisms. One major model improvement compared to previous studies was using variable and more realistic ozone deposition, a parameter, which controls the Br₂ emission yield from snowpacks in the model.

The model validation and relevance for the pan-Arctic region is unfortunately limited, because model predicted Br₂ emissions are strongest mostly outside the area covered by the observations used (e.g. windy hotspots or some of the coastal areas). This limitation should be mentioned in abstract and conclusion. Using in addition BrO from satellites as in Huang et al. (2020) would have allowed to validate the model for the wider Arctic region.

The manuscript requires some revisions, which are listed below.

SPECIFIC COMMENTS

I21 reactions on wind blown snow and on aerosol (e.g. Hara et al., 2018)

I33-34 This is somehow not complete as SSA from blowing snow does not only provide additional surface area for heterogeneous halogen chemistry but also additional bromide. Please clarify and add detail how aerosol is treated in the model as a finite bromide reservoir. How does modelled bromide depletion in aerosol compare to observations?

I34-36 This statement refers to the model calculation, which cannot be corroborated for the entire region by comparison with the observations used in this study (see above general comment). Please clarify.

I97 how is pH of the heterogeneous phase (snow, aerosol) treated in the model?

I201-202 Is it reasonable to assume the bottom 200m are well mixed during the period considered considering that surface inversions occur typically in winter/early spring? Please expand.

I220-21 Heterogeneous chemical reactions can convert SSA-transported bromide into gaseous reactive bromine species in the atmosphere. How is this modelled? What pH is assumed/calculated for open ocean and blowing snow SSA?

I232--233 even if all thresholds are met there needs to be snow on sea ice present to get airborne, thus this is a potential SSA production rate and therefore an upper limit. Please clarify.

I245 great to use high resolution wind speed data ($0.5 \times 0.625^\circ$) to capture wind gusts, relevant to both BSn and open ocean SSA!

I266--272 rather than choosing a single value based on a few point measurements it seems more sensible to explore the sensitivity of this important parameter with a few sensitivity model runs; same applies for SSA/SP ratio (I277) and the Br- enrichment factor

I345-47 how did you decide on 10cm snow depth threshold? shallow snowpacks near the coast may contain enough Br- for significant halogen activation; how much area is

affected by this filter?

I396-97 Please elaborate how the calculation of cloud pH is improved. Given that the multiphase reaction of bromide to reactive bromine depends on acidity explain also how snow pH and aerosol pH are computed or assumed in the model (see previous comment).

I470-71 this sentence is in contradiction to the previous that O₃ dry deposition should be lower above ice covered ocean than above open ocean. Please clarify.

I552-53 Please include a table and list a quantitative measure of model skill (e.g. root mean square error). I am not sure why only May is discussed here, I would like to see also the other months. Thus do a month-wise comparison between observations and model using the hourly data Feb-June in Table and include figures as Fig6 also for ea month, possibly in the appendix.

Section 4.1 Overall it appears the Br₂ yield from snowpacks is limited by surface resistance and ozone deposition and not availability of sunlight. Can you comment?

I628 Section 4.2 It would be very informative and strengthen the paper if the modelled O₃ was compared also to O-Buoys data. O₃ is measured by the O-Buoy platforms, so are data not available for the time periods considered?

I706 not only modeling should be done but also snow sampling and analysis as surely there are no or few data to back up your speculations.

I722-23 match within the uncertainty? This is not very quantitative, use a quantitative measure of model skill throughout as suggested above.

I727 "We extend our model run to the full year and find that enhanced daytime Br₂ yield can lead to increased Arctic Ocean Br₂ production in the summer" But this in disagreement with observations, as there are no ODEs in summer, please explain.

Fig1/I70-71: I find the figure confusing, at least use colour for heterogeneous reaction arrows (instead of dashed line) and associated text. I'd also suggest to include the Supplemental Table S1 into the main text to better follow the discussion and have a reference for each reaction right next to Fig1.

Fig2, check caption: MODIS image is the main figure, inset map shows the image footprint
FigS4 - show also for O-Buoy position (extracted from the model run) and discuss

TECHNICAL CORRECTIONS

l224-25 grammar. drop "which"?

l450-51 is this part of Fig4 caption? please remove separating line (similar for most captions)

l598 ozone deposition?

l680 PACK?? please check

all FIGURES' reproduction is fuzzy, use vector graphic.

REFERENCES

Hara, K. et al.: Important contributions of sea-salt aerosols to atmospheric bromine cycle in the Antarctic coasts, *Scientific Reports*, 8(1), doi:10.1038/s41598-018-32287-4, 2018.

Marelle, L., et al.: Implementation and Impacts of Surface and Blowing Snow Sources of Arctic Bromine Activation Within WRFâChem 4.1.1, *J. Adv. Model. Earth Syst.*, 13(8), doi:10.1029/2020ms002391, 2021.