Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., author comment AC3
https://doi.org/10.5194/acp-2022-44-AC3, 2022

Reply on RC2

William F. Swanson et al.

Author 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-AC3, 2022

Response to Reviewer 2
SPECIFIC COMMENTS
l21 reactions on wind blown snow and on aerosol (e.g. Hara et al., 2018)

We have added a mention of aerosols as possible reaction source and cited Hara et al. (2018). Furthermore, we have standardized our language throughout the paper and in our title to specify that we use a blowing snow SSA mechanism rather than a blowing snow mechanism, as Hara et al. (2018) and others make a convincing case that the blowing snow itself is not the primary source of increased reactive bromine.

l33-34 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?

I have clarified in the introduction to specify that the bromide on those aerosols is the direct source of increased BrO. We follow the same bromide treatment detailed in Huang et al. (2020), where they examine not only the depletion of aerosol but the deposition of bromide onto surface snow. They found that their predicted surface snow enrichment was consistent with observations of atmospheric chemical deposition to surface snow at Alert from Macdonald et al. (2017).

l34-36 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.

These lines are only intended to report on model predictions driven by the snowpack and blowing snow sources. I have specified that these lines refer only to modeled BrO.

l97 how is pH of the heterogeneous phase (snow, aerosol) treated in the model?

Aerosol is emitted within GEOS-Chem as alkaline based on the salinity of seawater. When this alkalinity is titrated out, the aerosol is considered to have a pH of 5. I included a description of the alkalinity titration in Section 2.3.

l201-202 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.

The BrOpptv200 quantity derived by assuming that the lowest 200 m are well mixed is not used in this paper. This quantity is included in our datasets, but as its derivation is described in the metadata we have eliminated these unnecessary lines. We are aware that is not reasonable to assume that the entire lowest 200 m of the Arctic troposphere are well mixed; for example, the OASIS field campaign measured mixing layer heights of 50 m during ozone depletion events (Boylan et al., 2014). The BrOpptv200 quantity is of some use as a rough comparison between predicted surface mixing ratios and MAX-DOAS measurements, but the method for direct comparison detailed in Section 2.7 should be used for more rigorous model comparison to MAX-DOAS measurements.

l220-21 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?

This is modeled as a heterogenous reaction on the aerosol surface. The pH of the SSA is based on the composition of seawater, which grants the aerosol a specific alkalinity. This alkalinity can be titrated within GEOS-Chem by deposition of acidic gases to a minimum of pH=5 (Alexander et al., 2005).

l232--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.

We clarify our previously unstated assumption that snowpack exists on all sea ice surfaces during the Arctic Spring. We believe that this is a reasonable assumption even on first-year sea ice freshly formed in fall due to the accumulation of snow over the winter months.

l245 great to use high resolution wind speed data (0.5x0.625º) to capture wind gusts, relevant to both BSn and open ocean SSA!

We find these high-resolution SSA datasets very useful for reducing model runtime and generating resolution-independent results. These results may not be appropriate for all applications because they rely on specific constant values chosen in the blowing snow SSA mechanism (see Section 2.4).

l266--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 (l277) and the Br- enrichment factor
The computational expense of running a full year at high resolution is high, and follow the findings of Huang et al. (2020) in which the performed several sensitivity studies over in March and April and settled on the parameters used here based on comparisons to satellite BrO observations.
l345-47 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?

While snow with a depth of less than 10 cm may be able to recycle snow, it will be less effective at producing reactive bromine that deeper snow. We decided on the 10 cm snow depth threshold based on detailed snowpack modeling studies, especially Thomas et al. (2011) predicting active bromine chemistry in interstitial air at 10 cm or below. The 10 cm snow depth screen mainly affects grid cells in more southerly latitudes and has little effect on Arctic production. Test Figure 1 shows the cells with snow affected by the 10 cm depth screen on May 1 2015. The perimeter of colored grid cells with snow depth less than 10 cm is further south earlier in the year due to cold winter temperatures.

Test Figure 1 Details: Snow depth in MERRA-2. Grid cells shown in bright yellow have snow depth greater than 0.1 m, cells shown in pink and orange represent MERRA-2 grid cells with a snow depth greater than zero and less than 0.1 m that are eliminated by our screen. White grid cells have zero land snow depth in MERRA-2 files.

l396-97 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).

We have specified how SSA pH is dependent on titration of aerosol alkalinity in Section 2.3. Shah et al. (2020) increased minimum background HCOOH, performed sensitivity analysis, and adjusted key parameters such as aerosol mass partitioning, gas-water equilibration time to improve the accuracy of GEOS-Chem cloud pH calculation. We would like to direct readers to Shah et al. (2020) for more detail on adjustments to cloud pH calculations within GEOS-Chem.

l470-71 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.

The Toyota 2016 paper only does direct comparisons between bulk Richardson numbers over sea ice and open ocean, finding that the air over sea ice is more likely to be stable. While they do discuss deposition velocities, their discussion is in terms of the sensitivity of ozone deposition velocity over ice to the choice of stability correction algorithm and does not make any blanket statements about the relative deposition velocities over sea ice and open ocean.
I have removed the first sentence, as their discussion of deposition velocity calculation is of interest for deposition scheme design but not relevant to this paper.

l552-53 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 each month, possibly in the appendix.

We have calculated RMSE for BrO on PACK, BLOW, BLOW+PACK, PHOTOPACK, and BLOW+PHOTOPACK in Table 2. We discuss these model skill measurements in Section 4.1.
We choose to highlight May here as it has the best data coverage for visual inspection. Our O-Buoy observations start on April 22 and end by June 10, an additional 17 days outside of May. A discussion of monthly O-Buoy accuracy would be skewed by number of valid observations. We have included graphs of hourly BrO predictions vs observations for the entire spring season as supplemental figures S7 and S8.

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?

We note that ozone deposition makes up over half of our Br2 yield. Availability of sunlight is not generally an issue, as the sun is up for the majority of the day over the Arctic Ocean after the vernal equinox. The sunlight scheme does increase the ozone yield 75-fold, so while sunlight is not the limiting factor it plays an important role in the PHOTOPACK runs.

l628 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?

O-Buoy 10 did not collect any ozone data in 2015, and O-Buoy 12 did not have any overlapping ozone and BrO observations. The ozone data that was gathered by O-Buoys in Spring 2015 has a number of data gaps, and is more difficult to analyze than the reliable BARC station ozone data. I have included graphs of ozone on O-Buoys 11 and 12 in the supplement as Figures S10 and S11.

l706 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.

We have been told by experimental groups that fall sampling of snowpack is difficult due to the fractious nature of the ice at this time, making snow sampling methodology difficult. We have included a call for fall sampling and analysis in the manuscript to encourage such an effort.

l722-23 match within the uncertainty? This is not very quantitative, use a quantitative measure of model skill throughout as suggested above.

Our results shown here match within measurement error. Table 2 calculates model RMSE for O-Buoys 10, 11, 12 and at Utqiagvik and discusses these numbers in Section 4.1.

l727 "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.

We note in Section 3.1 lines 474-475 that satellites see minimum BrO over that Arctic ocean in summer, which is at odds with the model predictions using an enhanced daytime Br2 yield. We agree with your statement, and we hope that modelers reading this paper will consider this when deciding upon their snowpack Br2 production schemes.

Fig1/l70-71: 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.
We have moved Supplemental Table S1 to Section 4 in the main text as Table 2 for clarity. We have referenced this table in the caption for Figure 1.
Fig2, check caption: MODIS image is the main figure, inset map shows the image footprint

We have corrected this caption.

FigS4 - show also for O-Buoy position (extracted from the model run) and discuss

We have used the highly reliable data from sensors at BARC for this graph. The O-Buoy windspeed sensors have data gaps due freezing over, and we feel that the high-quality BARC sensor data does the best job at illustrating the periodic nature of BLOW SSA production.

TECHNICAL CORRECTIONS
l224-25 grammar. drop "which"?
Done

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

l598 ozone deposition?

Updated to deposition from depletion.

l680 PACK?? please check

Good catch, old SNOW run name has been replaced with current PACK term.

all FIGURES' reproduction is fuzzy, use vector graphic.

I have prepared this manuscript in Word. I have prepared most of these figures at 300 dp, but I believe that word compresses them when converting to PDF. I have replaced several fuzzy figures with cleaner figures, and I will contact the editor to supply them with original images for final publication as needed.

Papers cited in response to reviewer:

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Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2022-44/acp-2022-44-AC3-supplement.pdf