The authors' revisions have significantly improved this manuscript focused on the first observations of atmospheric BrCN. The majority of the reviewer comments have been addressed. In particular, the expanded abstract and many figures added to the SI are significant improvements. In addition, critical sampling and measurements details have been added to the Methods section. The remaining comments mainly focus on clarifications and revisions needed to the added/revised text. I refer below to line numbers in the ATC version of the manuscript.

We thank the reviewer for their thoughtful comments and we have the following responses (noted in different font), and have made changes to the manuscript (noted in red).

L227-236 (new text): The authors added the statement: "Losses of BrO on the actual I-ToF CIMS inlet were not apparent during calibrations that were performed after both ATom-3 and -4 deployments." More information is needed here about the calibrations, including reporting the calibration factors and the actual loss %s observed. The assertion that their inlet did not experience losses based on Liao et al. (2011), who used a different inlet, is not acceptable. They base this on both being operated "at a high flow rate" (L233 & 235). On comparison, the Liao et al. inlet had a flow rate of 900 slpm through a 7.6 cm ID aluminum pipe, followed by 13 lpm through a 25 cm, 0.65 cm ID Teflon tube. Therefore, this inlet is very different from the authors' aircraft inlet that was 0.75 m, 0.75cm ID PFA Teflon tubing at 6 slpm (L169-170). From personal experience using the Liao et al. and other inlets, we have observed major BrO losses (far greater than Br2/HOBr problems) when the high flow through the large pipe is not used – and when only smaller diameter perfluoropolymer tubing is used. Further, the authors argument is inconsistent in that the inlet described by Liao et al. has been used for quantitative Br2 and HOBr measurements (including by coauthor – Wang et al. (2019) PNAS), whereas the authors assert that their inlet is not reliable for Br2 and HOBr (L236-237), showing that their inlet experiences increased losses compared to the design described by Liao et al. Therefore, I urge the authors to: 1) remove the added text on L229-234 that justify minor losses on their inlet based on Liao et al., 2) add %s BrO loss measured (and information about that test), and 3) add a statement that losses of BrO may have occurred on the inlet.

There are two factors at work in the inlet described by Liao et al (2011): initiation of flow from a stagnant airmass into a fast flow inlet, and behavior of BrO once it is in a Teflon inlet that is sampling off of that fast flow inlet.

As described by Liao et al., 2011 initiation of flow next to a structure requires a shroud and a very large flow in order to avoid sampling air that has had significant contact with surfaces of the structure. Even with this configuration, it's really the air that is in the core of the flow that is unperturbed by surface losses. It is therefore unsurprising that the Reviewer noted significant

BrO losses when the outer inlet was not used. The aircraft inlet used in our work and in the work of Liao et al., 2012a have no such flow initiation problems, as the aircraft was traveling at 100 m/s, or faster, and the Teflon inlet extended into that flow.

This leaves us with issue of possible losses of BrO on the Teflon inlets. The materials of the two inlets are essentially the same, but the residence times are somewhat different, which led us to use the experience of Liao et al. 2011 as evidence for our inlet. However, we have looked into the details of our laboratory tests on BrO loss and can now present quantitative information to demonstrate that our inlet does not have significant BrO loss.

The issues with $Br_2$ and HOBr that have been noted with our inlet are due to interconversion of these species especially in the presence of surface $Br^-$ ions. This is an effect quite independent of BrO chemistry, as that is either radical-radical chemistry or BrO aqueous-phase chemistry(Liao et al., 2012a), and the tests that we have done in between and after ATom deployments indicated no significant losses of BrO. This independence of BrO from $Br_2$ and HOBr chemistry is in agreement with previously reported tests on a PFA inlet that was slightly longer and of lower flow than our inlet used on ATom (Liao et al., 2012a).

As a result of this discussion about flow initiation, the similarity of the Teflon inlets between our study and (Liao et al., 2012a; Liao et al., 2011), and the results of our BrO tests: 1) we feel some modification, not elimination, of the text in Lines 229-234 is warranted, 2) we can now add that at least 90 ±4% of BrO is transmitted through the ATom inlet, 3) we prefer to state "we have not observed evidence of any other losses of BrO on the ATom inlet".

The added/modified sentences from Lines 228-236 are:

Significant losses of BrO on the actual I-ToF CIMS inlet were not apparent during calibrations that were performed before and after both ATom-3 and -4 deployments. In addition, the intercomparison of *in situ* BrO measurements by CIMS with Long Path Differential Absorption Spectroscopy (LP-DOAS) reported by (Liao et al., 2011), and the tests reported by Liao et al., 2012a provide further evidence that once flow into a Teflon inlet is properly initiated, an inlet of our type would not have significant BrO losses. Aside from a large shroud that was required to establish flow from a static air mass (Liao et al., 2012b), (not required for an aircraft inlet) the key feature of the Liao et al. (2011) CIMS inlet was a short section PFA Teflon tubing heated to 40°C and operated at high flow rates. Liao et al., (2011) report excellent agreement between the two methods for periods of steady wind flow without local NO pollution. Our inlet is slightly longer than that CIMS inlet, but was made of the same material and operated at a high flow rate.

Laboratory tests were conducted during ATom calibrations to assess the amount of loss of BrO in the instrument inlet used in ATom. Those tests compared the signals observed when adding the BrO source directly to the IMR, with those observed with the addition of the 0.7m long ATom inlet operated at 6.3 SLPM. The results showed the ATom inlet transmitted 90 ±4% of BrO, leading us to conclude there was minimal loss of BrO. We further note that this inlet loss was accounted for in the applied calibration factor, determined with the ATom inlet in place. We have not observed evidence of any other losses of BrO on the ATom inlet.

L226-228: The authors cite Neuman et al. (2010) and Osthoff et al. (2008) for the calibration of the I-ToF CIMS, but those prior papers report

calibration of the quad-CIMS. While the approach can be the same, the calibration results are expected to be different between the two CIMS instruments and also are expected to vary based on H2O addition and potentially RH, which needs to be addressed.

The reviewer is correct that these previous papers involved Quad -CIMS instruments with different IMR configurations, and we see by the way this sentence is worded that the reviewer could be mis-led into thinking that calibration factors from these references were used somehow in the current work, when really it is only the calibrations methods (i.e. sample preparation and independent determinations) that were used to calibrate the ToF-CIMS. Therefore, none of the requested information on the quad instruments is relevant. Instead, we will rewrite this sentence to make it clear we are referring to the methods of how the ToF-CIMS was calibrated and include further references about that. The sentence will now read:

Methods for the calibration of the I-CIMS for those species have been described previously(Neuman et al., 2010; Osthoff et al., 2008; Wild et al., 2014) and were applied to the calibration of the ToF-CIMS used in this work.

L175-180 (new text): Please report the measured scrubber efficiency (%) for the tested compounds – Br2, Cl2, and BrCl. Was the scrubbing efficiency measured for BrCN in the lab? I note that added Figure S2 is useful.

We have now added to the sentence on line 176:
and found to be more than 99% efficient at removing these dihalogens.

We did not check BrCN removal in the laboratory, but instead presented Figure S2, which essentially shows that the BrCN was removed at better than 92% efficiency. We have a sentence at lines 179 to read:
The signals during those zeroing periods indicate that the scrubber was at least 92% efficient at removing BrCN.

L379 (new text): Raso et al. (2017, PNAS) showed Br2 and I2 snowpack production, but I2 was not measured by Custard et al. (who also measured snowpack BrCl production, which may be relevant here) or Pratt et al., cited here. Please add Raso et al. for the mention of I2.

We have added Raso et al., 2017 to this list of references.

Minor Comments:
Line 101 (new text): Fix year (reference listed as published in 2077!).

The date on this reference should be 2007 and has now been fixed.

Line 287: NOy includes NOx, so this header should be corrected to NOy

instead of NOxy.

We are sorry for the confusion. The term 'NOxy' is our short-hand for this instrument, so its use here is inappropriate. The heading for the section is now changed to:

$NO_y$, $NO_x$ and $O_3$ measurements.

Line 572 (new text): Fix typo and clarify sentence.

We have fixed the typos and added some clarifying text. The sentence now reads:

Higher $Cl^-$ and $Br^-$ concentrations will serve to shift the pH at which BrCN becomes competitive to higher values, but the range still appears to be 6.5-8.5 pH units, a range that is applicable to matrices in polar environments.

Line 699 (new text): Cite Custard et al. (2017, ACS Earth & Space Chem) here for this statement, since it is not a result of this study.

We have now cited Custard et al., 2017 at this point.

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