We thank the reviewer for their in-depth thoughts and comments. As you will see below that we responded carefully to all your questions raised. In the text below, the reviewer's comments are shown in black, and our replies shown in blue and italic.

This manuscript describes interesting measurements of snowpack composition including salinity, bromide, and nitrate. Net deposition trends are observed, as well as profiles of ionic species in the snowpack and small-length scale differences in deposition around the Eureka area. However, the authors continue to try to say their data show that "the release flux of reactive bromine from snow must be a weak process and smaller than the derived bromide deposition flux of ~1×10^7 molecules cm-2 s-1, which flux is smaller than previously estimated flux by a factor of more than an order of magnitude." I believe the arguments they make on this point are flawed, as described below. I agree with the prior reviews that this manuscript is difficult to read and doesn't present a clear story, also making it hard to understand some of their arguments. I would suggest they clarify their description of observations and unless they can make a valid argument that their data actually constrains short-timescale snowpack emissions, remove that point.

Response: As you will see below that we carefully further examined our major conclusions by showing new evidence, however, we do agree with the reviewer that the method applied in this study could not fully resolve short-timescale snowpack emissions. Thus, the flux derived only represents an AVERAGE flux over a long-term period (~1 month).

Snow sampling and small flux challenges:

Fundamentally, the low concentration of bromide in the surface snow along with variability in ion concentrations related to sampling different surface snowpack (your next day's snow sample will be to the side of the prior day so you don't dig a hole) means that it is difficult to quantify changes in snow composition over time. The authors attempt to make up for this inherent challenge by using a fairly long timeseries. This approach means that the NET long-term deposition trend is quantified. Prior reviews pointed out that analysis errors and variability make it hard to quantify the net deposition trend, and the response was that "...collecting daily snow samples over a relatively long period of 3-4 weeks with an aim of detecting the possible accumulative change." This clearly shows that the study authors understand that they only detect the "accumulative change" = NET deposition.

Response: Yes, the measured flux is a NET deposition flux.

Net deposition does not constrain short-term snowpack emissions:

Net deposition is measured, but there may be larger fluxes that are bidirectional (snowpack emission and deposition) occurring on shorter timescales. Therefore, the authors cannot say that the long-term trend in NET deposition constrains shorter-term fluxes to be small. If the shorter-term fluxes were unidirectional only (e.g., deposition only), then the net deposition can constrain the process (there is no emission in this presumed unidirectional case), but in the case of bromine, we know that snowpack can both emit reactive halogens and have halogens deposit to it. The short term (days to hours) variability in both BrO and snow Br- also are consistent with significant short-term fluxes.

Response: Note that the small net deposition flux measured could be attributed to two different scenarios: (1) the emission and the deposition fluxes are both large and the net flux is a residual of the two fluxes; (2) the net flux is determined mainly by the deposition flux, and the emission flux is relatively small and thus can be ignored. Obviously, scenario 1 is what the reviewer suggested, while scenario 2 is what we assumed. Note, the measured short-term (days to hours) variability in both BrO and snow Br- does not necessarily lead to snowpack emissions, as other mechanisms such as blowing snow can result in a similar episodic perturbation.

New "Bromine mass balance" approach (section 3.7):

The authors make an equation for the time trend in air column density, equation R7, which says dc_air / dt = P_air - c_air / tau_air. They go on to say: "However, from Figures 5(c) and 6(c), we see a significant decreasing tend of BrO partial column, indicating the input term P_air is much smaller than the loss term, c_air / tau_air."

This doesn't make mathematical sense. For equation 7's left side to be negative, P_air should be smaller than c_air / tau_air, but a large value of P_air can be allowed as long as c_air / tau_air is larger.

As an example, let's say that there is no trend in gas-phase BrO (steady state), then the left side of R7 is zero, which means that 0 = P_air - c_air / tau_air, which gives the steady-state result: P_air = c_air / tau_air. Let's take tau_air to be 1 day = 86400s and say BrO is 3e13 molecule cm^-2 (typical value from their plots), and they assume BrO is 0.3 of gas phase Br, so gas-phase c_air = 1e14 molecule cm^-2. Then the production rate is P_air = 1e14 molecule cm^-2 / 86400s = ~1e9 molecule cm^-2 s^-1. This emission flux is within the quoted measurement (in the literature) of "snowpack bromine emission, a direct gradient measurement of Br2 and BrCl above a patch of snowpack was made near Utqiaġvik, Alaska (Custard et al., 2017), who reported emission fluxes of 0.7–12 × 10^8 molecules cm−2 s−1." Instead, the authors decided to choose a lifetime of reactive bromine of 42 days in 2019, and due to this choice, they get a much smaller emission flux. This lifetime of reactive bromine seems unreasonably long given the episodic nature of reactive bromine events, and is discussed below.

If we simply look at the BrO timeseries in Figures 5c and 6c, we can see that BrO varies significantly during individual days. BrO doubles on some days, and there are many instances where there is a factor of two difference in BrO between one day and the next. If we interpret this variability as due to local fluxes, one would clearly accept that BrO lifetime can be on the order of a day, which would allow snowpack emission fluxes comparable to the measured result from Custard et al., 2017.

Response: As mentioned above there are two different possible scenarios (1 & 2) that could result in very small net deposition fluxes. If the system is bi-directional, then the derived flux (representing an average flux) underestimates the large short-term fluxes; however, if the system is unidirectional, then the derived net flux represents well the actual emission flux occurring.
Note that we did not arbitrarily choose a long "lifetime", the number of 42 days was derived from an exponential fit to the 2019 spring (March to May, not March) BrO data (see figure below and relevant text, which was not deliberately discussed in our previous response. The derived long "lifetime" should not be treated as the actual lifetime for reactive bromine as obtained in a more isolated air parcel. This is because the timescale was derived from long-term seasonal BrO data, where the system is open and affected by many factors, thus the long "lifetime" most reflects a seasonal decay of atmospheric bromine. Therefore, we used the term "seasonal decay lifetime" to distinguish it from the actual lifetime of reactive bromine.

Complications with their lifetime analysis:

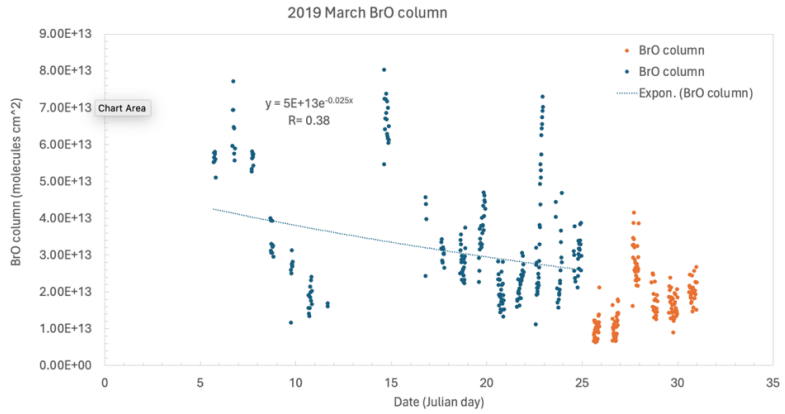
This study was done at a time when the reactive halogen season was declining a bit on a ~month timeframe. This slight decline in net BrO is calculated to be a loss of c_air over time in their equation 7. They then go on to use the same BrO data with an exponential fit to calculate a "loss rate" of reactive bromine. Use of the same data on both sides of equation 7 appears circular.

Response: We think the reviewer might have misunderstood the method used to derive these two parameters. The mathematical solution of equation R7 is an exponential function, which can be rewritten as a linear function as long as the timescale of Tau_air (or "lifetime", =42 days) is much larger than 1 day. Therefore, the loss rates derived directly from the linear fit and from the exponential fit to the BrO data should give very similar results. With an exponential fit, one can work out precisely the loss rate at any given time, but they are not a mean loss rate, to get a mean loss rate for a period we need to do integration and averaging. To avoid this process, we can directly apply a linear fit to the same dataset (as we did in this study). Therefore, we did not apply the same data on both sides of R7, we only applied the same BrO data to derive the two parameters.

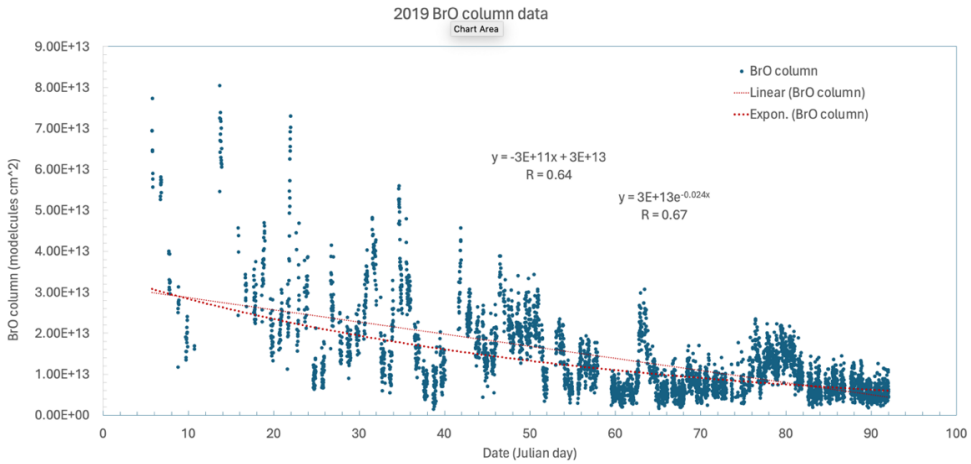
Note that this "loss rate" is a NET loss rate over many weeks, not a loss rate that is specific to shorter term processes. Let's for the sake of argument say that they had stopped their study six days earlier in 2019. It appears that the trend in BrO over time would now be increasing, and if you fitted it to an exponential, you would not have a loss rate, but a growth rate or possibly flat (zero slope, infinite lifetime). They take the exponential loss rate to be be indicative of the lifetime (tau_air) of reactive bromine, but now the loss rate would be very small and the "lifetime" of reactive bromine would be longer than the 42 days they calculate in 2019 -- now reactive bromine might live well into the summer or even over multiple years, which is counter to observations. Is is obvious that you cannot extract the lifetime of reactive bromine in the way they are trying to

do it here. Without a constraint on tau_air, they cannot use equation 7 to calculate the magnitudes of the two terms on the right side, and thus they cannot determine the snowpack production flux.

Response: The reviewer did raise a key issue regarding how to precisely constrain lifetime (or tau_air) and loss rate from the field data. Following referee's suggestion (removing the last six days from the March BrO data), we did perform the experiment with the result shown below. It gave a Tau_air of 40 days with a small R=0.38. When the whole March data are considered, the significance increases but the uncertainty remains large. In this study, instead of using the March BrO data, we took the whole spring season (Math to May) BrO into consideration. As shown below in new Figure S6 (in the supplementary material), BrO has a decline trend from March to May, which trend is statistically significant (R> 0.6). Using the regression fits we derived the 42 days "lifetime" and loss rate of 3E11 molecules cm2 d-1. As mentioned above this long timescale "lifetime" is different from the actual short lifetime of reactive bromine obtained in an isolated airmass, it more likely represents a seasonal decay of atmospheric bromine species, affected by many other factors involving both physical and photochemical processes. The decline of BrO from early spring to late spring was also exhibited in other years (from 2016 to 2018) at Eureka as shown in Bognar et al. (2020), which demands further investigation.



This figure shows the 2019 March BrO column data, in which an exponential fit to the data (excluding the last six days) was used to derive lifetime, as suggested by the reviewer.



New Figure S6. 2019 BrO column data from March 5th to May 31st, in which the exponential fit and the linear fit are inserted. Note that Julian day (X-axis) was introduced in this figure for the regression fits.

To show that equation 7 cannot be used in this manner, consider this hypothetical situation. Say there was a sealed test tube with some liquid water and vapor in it. We now raise and lower the temperature, which will cause water to evaporate (P_air) and raise c_air or condense and lower c_air. Over a long time (many warming and cooling cycles), if you fit the timeseries of c_air to a slope, dc_air/dt would be close to zero (much like their long-term trend in gaseous bromine is fairly flat). By their method, they would then fit the c_air over time to an exponential, and say that the lifetime of the vapor is long (because the timeseries is on average flat), so tau_air

is very long, and the second term (c_air / tau_air) will go to near zero. Equation 7 will then be dc_air/dt = ~0 = P_air - c_air/tau_air (term = ~0), so you find P_air = ~0. Their interpretation is that this system has no evaporation of water (P_air = ~0), while in fact water is evaporating and condensing with potentially large fluxes. The failure is that you cannot use the net flux to constrain faster bi-directional fluxes.

Realistically, over multi day periods, the weather changes, airmass origins change, the sun rises, temperature warms on average. These factors lead to wide variability in BrO as shown on their figures. Yet, they fit a long-term trend through the data and call that the lifetime of reactive bromine as if there were a constant loss rate for the full campaign, nearly a month. It is clear that faster than monthly processes are needed to describe reactive halogen chemistry and that long-term "accumulative changes" do not directly constrain faster underlying bi-directional fluxes.

Response: Unfortunately, we could not fully agree with the referee on this topic. The derived near-zero evaporation flux from the suggested experiment is "reasonable" in our view, this is because the tube is a sealed system and there should not be a net evaporation flux as a long-term mean. Here we suggested another similar experiment by replacing liquid water and water vapour in the tube by mercury and mercury vapour. Since mercury, compared to water, is less sensitive to temperature change, there will be no such large short-term fluxes during the experiment period, thus a near zero evaporation flux and a large lifetime number will be derived, but they are completely correct. Therefore, the key issue is not in the method itself, the interpretation of the results relies on whether there are short-term bi-directional fluxes in the system.
We agree with the referee that the method used here could not resolve short-term flux (if there are such short-term fluxes), thus in the revised version, we used "average" flux in our statements. For instance, in section 4 on discussion, we stated: "the method applied could not resolve short-term (<1 day) fluxes, therefore the derived fluxes only represent an average flux over the campaign period (3-4 weeks)."

Summary:

Overall, the central problem with this manuscript is that the authors do not accept the difference between a net deposition rate measured over a month-long period and faster bi-directional fluxes that are occurring (based upon prior literature reports of snowpack emissions). They cannot constrain fast fluxes that happen in bi-directional manners by a long-term deposition flux. The BrO measurements, which vary by factors of two day to day could clearly be consistent with large snowpack emission on one day followed by deposition back to the snow the next day. Alternatively, look at the snowpack Br- timeseries, which shows a lot of variability. It is clear that analysis errors and snow sampling variability can affect variability in measured Br-, but one were to believe that this variability were real, it would indicate large fluxes of reactive bromine out of the snow and re-deposition of Br- back to the snow.

Over the long-term, due to bi-directional fluxes, the net change in snowpack bromine could be small (as is observed), but a lot of chemistry could have happened on shorter timescales than their long-term net trends can capture. Effectively, the approach described in this manuscript is not equipped to put short-term constraints on snowpack emissions fluxes.

If the authors want to report snowpack composition, vertical profiles in pits, and net deposition fluxes over long periods of time, I can see the publication of a manuscript showing those results. However, I see no validity in the attempts they have presented to constrain short-term snowpack emissions fluxes. If the authors seek to maintain that point, I argue for rejection of the manuscript. In this set of comments, numerical examples derived from their figures were shown to be consistent with snowpack emissions fluxes measured by others and reasonable lifetimes for reactive bromine. The lifetimes and fluxes of these faster processes fit with variability observed in both atmospheric reactive bromine and snowpack bromide.

Response: We accepted the reviewer's key point that our method could not resolve faster short-term bi-directional fluxes, however, we defended our major conclusion of the weak snowpack emission flux, though this flux only represents a mean flux over the period of ~1 month. In section 5 we stated: "Through the mass balance analysis we conclude that the average emission flux from snow over the campaign period should be less than the average bromide deposition flux of ~1×10^7 molecules cm-2 s-1, which is an order of magnitude smaller than previously measured emission flux of 0.7–12 × 10^8 molecules cm−2 s−1 (Custard et al., 2017). Note that the net mean fluxes observed do not completely rule out larger bidirectional fluxes over shorter time

scales."