

Voss et al., A novel, balloon-borne UV/visible spectrometer for direct sun measurements of stratospheric bromine

Answers to Reviewer 1

The reviewer's comments are in black, *answers are in blue*.

This is a well-written paper that presents the design and first results from a new balloon spectrometer for solar occultation measurements of BrO. The instrumentation is described in detail and the co-authors present improvements to the standard retrieval approach together with a convincing handling of instrumental artifacts. The inferred bromine load and its derived uncertainty shows the usefulness of these measurements for monitoring stratospheric bromine trends. The paper is a good fit for AMT and should be published after addressing these minor revisions:

1. *We thank the reviewer very much for the encouraging evaluation of our manuscript.*

The case that the 1-2 ppt difference quoted here is actually of scientific significance should be made. On the surface it seems small in light of the total budget of 20 ppt.

2. *We agree and the following sentence is added to the manuscript:
This small difference in total bromine may point to some inorganic bromine being scavenged during its transport from the lower into the middle stratosphere and may accordingly reduce the bromine-mediated loss in ozone (Sinnhuber and Folkins, 2006).*

The system level descriptions of the solar tracker and the spectrometer units are well done, but would be greatly helped with schematic level drawings of the designs (in addition to the picture, which is a bit hard to follow for one not familiar with the instrument).

3. *In response to the reviewer comment the following schematic drawing of the solar tracker is added to the manuscript.*

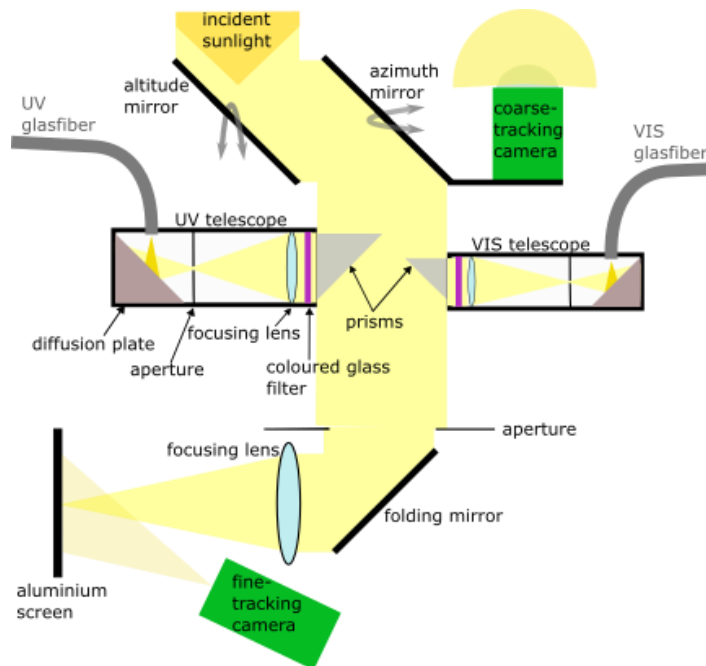


Figure 1: Schematic of the light path through the solar tracker.

How big is the non-linearity correction in the processing step, even when the detector saturation is kept to 30-60%?

4. *We added the following sentence to the manuscript.
The detector's response is most linear for pixel saturations between 30 % and 60 % with deviations from linearity within this range being smaller than 0.002 %. Thus, the correction in the processing step increases the signal of pixels with saturations between 30 % and 60 % by less than 0.002 %.*

Something is missing the explanation of how the dependence of etalon optical density on SCD is used for the retrieval process. How is the extrapolation to the larger SCDs performed throughout the retrieval range beyond the high sun range used for the characterization of the dependence?

5. *The sentence in the manuscript is changed to provide some details:
The inferred principal components are then used to correct all spectra including those used for scientific analysis. To this end, the DOAS retrieval includes a set of the inferred residual spectral structures as pseudo cross sections. The scaling of each pseudo cross section together with the gas absorption structures is determined via least-squares minimization.*

The significant difference in [BrO] for different polynomial degrees is somewhat concerning. Is there some level of non-orthogonality between the higher degree polynomials and the BrO cross section? If the residuals are the same, would it not make sense to take the result for the lowest degree polynomial as the most robust?

6. *We assume that the reviewer comment refers to Figure B1 and Figure C1 of the appendix. The polynomial referred to here is not the polynomial included in the DOAS fit itself. Rather the polynomial referred to is the polynomial fitted to the optical density time series to model τ_{atmo} of each pixel within the preprocessing procedure used to infer $\tau_{residual}$. (We changed the notation from τ_{etalon} to $\tau_{residual}$ in response to the second reviewer's comments.)*

The polynomial represents the increasing optical density of τ_{atmo} with increasing light path through the atmosphere with decreasing solar elevation. If τ_{atmo} would only increase due to Rayleigh scattering this would imply a linear function of the total SCD_{air} , however also strong absorbers such as ozone and possible instrumental dependencies on the solar elevation could change the measured optical density. Both effects may not scale linearly with SCD_{air} . Since both effects may also be wavelength dependent, the resulting (pseudo) extinction is different for different spectral pixels. Therefore, different polynomial degrees are fitted to τ_{meas} to model τ_{atmo} for each wavelength (pixel). By that, we can infer the residual optical densities. The mean quality of the polynomial fit to τ_{meas} (SCD_{air}) over all pixels is found to be similar for any of the chosen polynomial degrees of order 3 to 5. However, by visual inspection, it seems that for some pixels a polynomial of 3rd degree or lower does not capture the correct slope of τ_{atmo} (SCD_{air}) well, however a polynomial of 5th or higher order may additionally compensate for some variations in $\tau_{residual}$.

How will new CCD detectors avoid the contamination problem? It seems the cause is not well understood by the co-authors, so might it not happen again? Maybe it is from internal reflection in the focal plan array itself (this is a known phenomenon)?

7. *In past studies, both in the laboratory as well as in the field, we often used Ocean Optics QE-Pro spectrometers but did not encounter such problems of residual structures. Unfortunately, prior to the first deployment from Kiruna in 2021 a thermal incident occurred to the spectrometer unit due to an operating mistake with the current spectrometers in the laboratory. We accidentally left the Peltier detector cooling running while not providing cooling of the Peltier's warm back-side by the ice-water bath. Therefore, the spectrometer optics, electronic parts and housing heated up to 60°C while under vacuum. This resulted in outgassing of volatile chemical components within the vacuum chamber and accordingly their condensation onto the cold detector surface. For cleaning, we then reversed the process, heating the detector while cooling the lid of the vacuum chamber and evacuating the spectrometer container. By this measure, most of the condensate could be removed from the detector surface such that the residual structures were largely reduced. However, it is plausible that a residual layer of condensate remained on the detector surface causing residual optical structures with $\tau_{residual} < 10^{-3}$, which interfere with the retrieval of minor atmospheric absorbers. We are certain that what we called etalon structures is a result from this operating error and the contamination of the detector surface. Thus, we expect that by replacing the contaminated with new CCD detectors will eliminate this problem.*

In the revised version of the manuscript we added a short report on the thermal incident

*to the instrument performance section of the manuscript.
We changed the wording “etalon structures” to “residual structures” since we realized that the initial wording is too specific and misleading for the reader.*

Prolific use of brackets for parenthetical phrases and around quantities make the text a bit difficult to read. Check with the editor for guidance on correct typesetting.

8. The use of brackets was checked throughout the manuscript. Brackets used for parenthetical phrases were reduced by rephrasing the sentences. Square brackets around molecules are commonly used to indicate concentrations of these molecules.

Line 78: “correct for them”

9. The text is rephrased accordingly.

Line 88: rephrase “all electronics threatened to overheat”

10. The text is rephrased accordingly.

Line 155: rephrase “were more threatened”

11. The text is rephrased accordingly.

References:

Sinnhuber, B.-M., and I. Folkins, Estimating the contribution of bromoform to stratospheric bromine and its relation to dehydration in the tropical tropopause layer, Atmos. Chem. Phys., 6 (12), 4755–4761, doi:10.5194/acp-6-4755-2006, 2006.