

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

## Answers to Reviewer 2

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

In this manuscript, the authors describe a new balloon-borne DOAS instrument for measurements of stratospheric species. The instrument's performance during two deployments is discussed, and data from one measurement flight is used to estimate stratospheric Br<sub>y</sub> and compare the results to previous estimates.

The manuscript is well written and discusses an instrument and measurements relevant to atmospheric science and the study of the ozone layer. I understand that this article is intended as a reference for future work reporting on more measurement results achieved with this instrument. I recommend it for publication after the issues listed below have been addressed.

*We thank the reviewer for the appreciation of our work and the helpful comments. Please find our point-by-point reply below.*

### Major comments

As this is not the first stratospheric balloon-borne DOAS instrument, it would be good to highlight the actual progress of this instrument compared to the previous versions. This is not clear to the reader, and my impression is that while this instrument is new, it is not particularly innovative or better than past ones.

- 1. Previously, our balloon-borne DOAS instruments were flown together with the LPMA instrument which provided the solar tracker (Ferlemann et al. 1998, Weidner et al. 2005). Since the LPMA instrument and hence the solar tracker was no longer available, a new solar tracker was needed for balloon-borne solar occultation measurements. Fortunately, in parallel, a solar tracker was developed by our group to monitor greenhouse gases from the ground on slowly moving platforms (e.g. ground-based: Gisi et al. 2011; ship-borne: Klappenbach et al. 2015, on a pickup truck: Butz et al. 2017). The concept of this ground based solar tracker was adjusted for deployment on balloon gondolas. Compared to the former LPMA solar tracker (Camy-Peyret et al. 1995), the presented stand-alone solar tracker weights much less, is more compact and consumes less power.*

*Moreover, the concept and design of the spectrometer module was inherited from the limb spectrometers used by Weidner et al. (2005) and Kritten et al. (2010). However, in the present application the spectrometers were changed from Ocean Insight QE 2000 and QE65000 spectrometers to slightly larger, higher performance Ocean Insight QE-Pro spectrometers. Additionally, electronics including the onboard computer and power lines were updated. We expect that the new setup with better CCD detectors, in principle, offers higher sensitivity and stability than previously achieved. However, due to the spectral artefacts originating from detector contamination reported on in the revised manuscript, we*

*were not able to reduce the detection limits compared to previous balloon-borne DOAS experiments. In the future, after replacing the contaminated CCD detectors, we hope to reduce the uncertainties and to detect weaker absorbers such as IO. Compared to the LPMA/DOAS direct sun spectrometer, the new spectrometer weighs less while being more compact and thus can be placed flexibly on the gondola.*

*The presented, low weight and compact instrument consisting of both units is well suited to be deployed as secondary payload with several other instruments on the same payload.*

*The manuscript's instrument description has been updated with the information presented here.*

The description of the instrument is very detailed (I would say too detailed) in some parts, but it lacks drawings to understand the set-up of the telescope and the overall system. Figure 2 does not really help in this respect. Please add schematics providing more details on the set-up.

*2. A schematic of the solar tracker optics is added to the manuscript (see the above figure (Figure 1) added to comment #3 of reviewer 1).*

As discussed in the manuscript, there are two major problems with the current system: a) the sun-tracker is not fast enough to allow for measurements during ascent when the gondola is not stabilized, and b) there are artefacts in the spectra, which make BrO analysis difficult.

The first problem is only mentioned, and no indication is given regarding how it will be solved in future deployments. Not being able to measure profiles is a severe limitation of the instrument, so this needs to be discussed more.

*3. Yes, the reviewer is correct in his assessment.*

*The first limitation is mostly related to recent safety-related changes in the balloon flight train and modifications in the launch procedure. The operating agency (CNES) decided to replace the previously used auxiliary balloons, which were released shortly after launch, by one auxiliary balloon, that is no longer released and that has the flight train running through it. This change comes at the expense of a larger area of attack for the shear winds which induces strong torques to the flight train and the gondola. In consequence, a reliable azimuth stabilisation is not (yet) possible when strong shear winds are prevailing, i.e. during balloon ascent below altitudes of at least 10 km to 15 km. This hinders continuous ascent measurements due to the unpredictable rotations of the balloon gondola.*

*Of course, one could argue, a more rapid and improved sun-tracker, that is able to rotate freely by 360 degree without cable connections, could compensate for the deficit in the azimuth stabilisation of the gondola. However, it needs to be noted that although the sun tracker is mounted on top of the gondola its unobscured view to the sun is limited to an azimuth range of approximately 270° by the presence of the gondola structure, in particular the so-called pivot by which the gondola is azimuthally stabilized. The shading of the sun-tracker by these structures further complicates a fast and continuous tracking of the sun. The*

*realisation of such a tracker is a difficult engineering challenge which we did not pursue so far.*

*Some discussion on the above issue has been added to the manuscript.*

The second problem is discussed in great detail, but only concerning the approach taken to correct it in the spectra. Very little is said about the possible origins of the artefacts beyond the fact that they are linked to etaloning on the CCDs. I'm not entirely convinced by this explanation, as this appears to be a rapid process, and the question is, what would be condensing on the CCDs at this rate in an evacuated housing? In the sense of "what can the reader learn from this manuscript", I would hope for a more detailed discussion of possible sources and the tests performed to investigate them:

- 4. As described in detail in response #7 to reviewer 1, a thermal incident due to an operating error occurred prior to the Kiruna balloon launch contaminating the surface of the CCD detector. This mal-functioning manifested itself by etalon-like residual structures in the recorded spectra which we attributed to the presence of a condensate on the surface of CCD detectors. Follow-on attempts to remove the condensate on the CCD by temperature gradient assisted vacuum cleaning were partly successful in that we could reduce the residual structures due to condensate to a degree lower than the detection limit of the residual in the laboratory. However, for the spectra recorded during the balloon flights we assume some residual condensate remained on the CCD's surface.*

*A short report on the thermal incident is added to the instrument performance section of the manuscript.*

Has the effect been reproduced in the lab?

- 5. Several lab experiments were carried out to investigate if we can reproduce the artefact in the lab after the first deployment. However, we were not able to reproduce a similar behavior on the  $\tau_{\text{residual}} < 10^{-3}$  level, mainly because light sources (halogen lamp, mercury and krypton emission lamps) available in laboratory were less stable than the sun. Further using the sun in test measurement at the ground led to residual structures due to variability in atmospheric processes (absorption and scattering) of the order of the instrumental effect to be studied.*

Does it occur also if only the CCDs are cooled but not the rest of the optical system?

- 6. This was not directly tested, since in the lab experiments, the entire optical system was always kept at 0°C (to support the cooling of the warm side of the Peltier element) with the ice-water bath while the CCDs were usually cooled to -10°C by the thermo-electric cooler.*

Does it happen for both channels? Are the phases of the etalons on the two CCDs linked?

7. *Yes, it happened for the UV and visible spectrometer, but it was not linked in phase and frequency (periodicity) for both spectrometers. However, the periodicities were of similar length.*

Could it be etaloning due to condensation on another surface (mirrors, fibres, filters)?

8. *Due to the incident with the spectrometers described in response #7 to reviewer 1, it is rather likely that the artefacts are linked to the response of the spectrometers' detectors.*

Concerning the proposed correction approaches, I do not understand the rationale behind using SCD<sub>air</sub> as the axis – I would have thought that time is the relevant parameter here. Please explain.

9. *When describing the optical density changes in atmospheric spectra SCD<sub>air</sub> is the natural coordinate since Rayleigh scattering is the dominant process (e.g. Gurlit et al. 2005). And yes, very likely SCD<sub>air</sub> is not the natural coordinate to describe oscillations of the artefacts. Therefore, we investigated the oscillations of the spectral artefacts as a function of several parameters including time, SZA and SCD<sub>air</sub>, but the oscillations did not show a constant period as a function of any of these parameters. So, we decided to keep SCD<sub>air</sub> as the preferred coordinate.*

Clearly, the etaloning introduces substantial additional uncertainty even after correction, and this limits the value of the current measurements in addition to the limitation imposed by the lack of profiling capability.

10. *Yes, we concur. This is why we try to estimate the additional uncertainty by the sensitivity study. However, since the cause of this residual structure is not totally clear but did not show in previous deployments with similar spectrometers, we decided to replace the spectrometers for potential future deployments. With the replaced spectrometers, we expect the residual structures will no longer be present, permitting a lower detection limit with a lower error in the retrieved Br<sub>γ</sub>.*

### **Minor comments**

Figure 1: What is the sharp gradient in the green shading at 80.5°? This looks like an artefact.

11. *This is an artefact of the open street map tile. In the light of open science and open data, we would like to use the map provided by open street map nevertheless.*

Figure 3: The FWHM of the UV instrument is relatively small – is a convolution of the literature cross-sections necessary (and even possible) at this high spectral resolution?

12. *Yes, it is since the literature cross sections used for NO<sub>2</sub>, O<sub>3</sub> were recorded at much higher spectral resolution. The resolution of the literature O<sub>3</sub> spectra is about 0.06 nm, the literature NO<sub>2</sub> spectra is about 0.01 nm, and for the literature BrO cross section it is around the same spectral resolution as our instrument but recorded in 0.04 nm intervals. For O<sub>4</sub> the*

*absorption structure is very broadband, thus the convolution just maps the absorption structure to the wavelength grid of our spectrometer.*

L264: Calling the vertical column density VD is inconsistent with the other notation. Please use VCD<sub>air</sub>. Alternatively, if you meant density here (as suggested by your use of VD in equations 4 and 5), use another symbol such as  $\rho_{\text{air}}$  for air density.

*13. Air density is meant here, the notation in the manuscript is changed accordingly.*

L268: Why is it a good assumption that BrO is constant above floating altitude?

*14. A constant [BrO] within more than a scale height above balloon float altitude of 34 km is predicted by photochemical models. For example, the simulated SLIMCAT [BrO] varies by less than 5% between 34 km and 40 km altitude.*

Equation 4: This is not correct. Either you remove the  $\Delta h_i$ , or you replace VD with the concentration of air

*15. VD is supposed to be the density of air. For clearer notation, we changed VD to  $\rho$  as suggested.*

Equation 5: Same problem as for equation 4

*16. VD is supposed to be the density of air. For clearer notation, we changed VD to  $\rho$  as suggested.*

L291: predominant

*17. We changed the text accordingly.*

L461: I wouldn't say that NO<sub>2</sub> SCDs are unaffected, but they are not much affected.

*18. We concur and changed the text accordingly.*

#### References:

Butz, A., Bösch, H., Camy-Peyret, C., Chipperfield, M. P., Dorf, M., Dufour, G., Grunow, K., P., J., Köhl, S., Payan, S., Pepin, I., Pukite, J., Rozanov, A., von Savigny, C., Sioris, C., Wagner, T., Weidner, F., and Pfeilsticker, K.: Inter-comparison of Stratospheric O<sub>3</sub> and NO<sub>2</sub> abundances retrieved from balloon-borne direct sun observations and Envisat/SCIAMACHY limb measurements, *Atmos. Chem. Phys.*, 6, 1293–1314, 2006.

Butz, A., Dinger, A. S., Bobrowski, N., Kostinek, J., Fieber, L., Fischerkeller, C., Giuffrida, G. B., Hase, F., Klappenbach, F., Kuhn, J., Lübcke, P., Tirpitz, L., and Tu, Q.: Remote sensing of volcanic CO<sub>2</sub>, HF, HCl, SO<sub>2</sub>, and BrO in the downwind plume of Mt. Etna, *Atmospheric Measurement Techniques*, 10, 1–14, <https://doi.org/10.5194/amt-10-1-2017>, 2017.

Camy-Peyret, C., Jeseck, P., Hawat, T., Durry, G., Berubeé, G., Rochette, L., and Huguenin, D.: The LPMA balloon borne FTIR spectrometer for remote sensing of the atmospheric constituents, in: 12th ESA Symposium on Rocket and Balloon Programmes and Related Research, 1995.

Ferlemann, F., Camy-Peyret, C., Fitzenberger, R., Harder, H., Hawat, T., Osterkamp, H., Schneider, M., Perner, D., Platt, U., Vradelis, P., and Pfeilsticker, K.: Stratospheric BrO profiles measured at different latitudes and seasons: Instrument description, spectral analysis and profile retrieval, *Geophys. Res. Lett.*, 25, 3847–3850, 1998.

Gisi, M., Hase, F., Dohe, S., and Blumenstock, T.: Camtracker: a new camera controlled high precision solar tracker system for FTIR-spectrometers, *Atmos. Meas. Tech.*, 4, 47–54, <https://doi.org/10.5194/amt-4-47-2011>, 2011.

Gurlit, W., Bösch, H., Bovensmann, H., Burrows, J. P., Butz, A., Camy-Peyret, C., Dorf, M., Gerilowski, K., Lindner, A., Noël, S., Platt, U., Weidner, F., and Pfeilsticker, K.: The UV-A and visible solar irradiance spectrum: inter-comparison of absolutely calibrated, spectrally medium resolution solar irradiance spectra from balloon- and satellite-borne measurements, *Atmos. Chem. Phys.*, 5, 1879–1890, <https://doi.org/10.5194/acp-5-1879-2005>, 2005.

Klappenbach, F., Bertleff, M., Kostinek, J., Hase, F., Blumenstock, T., Agusti-Panareda, A., Razinger, M., and Butz, A.: Accurate mobile remote sensing of XCO<sub>2</sub> and XCH<sub>4</sub> latitudinal transects from aboard a research vessel, *Atmos. Meas. Tech.*, 8, 5023–5038, <https://doi.org/10.5194/amt-8-5023-2015>, 2015.

Kritten, L., Butz, A., Dorf, M., Deutschmann, T., Kühl, S., Prados-Roman, C., Pukite, J., Rozanov, A., Schofield, R., and Pfeilsticker, K.: Balloon-borne limb measurements of the diurnal variation of UV/vis absorbing radicals – a case study on NO<sub>2</sub> and O<sub>3</sub>, *Atmos. Meas. Techn.*, 3, 933 – 946, 2010.

Weidner, F., Bösch, H., Bovensmann, H., Burrows, J. P., Butz, A., Camy-Peyret, C., Dorf, M., Gerilowski, K., Gurlit, W., Platt, U., von Friedeburg, C., Wagner, T., and Pfeilsticker, K.: Balloon-borne limb profiling of UV/vis skylight radiances, O<sub>3</sub>, NO<sub>2</sub>, and BrO: technical set-up and validation of the method, *Atmos. Chem. Phys.*, 5, 1409–1422, <https://doi.org/10.5194/acp-5-1409-2005>, 2005.