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Interactive Comment

Interactive comment on "Balloon-borne stratospheric BrO measurements: comparison with Envisat/SCIAMACHY BrO limb profiles" *by* M. Dorf et al.

M. Dorf et al.

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[]letter times amsmath

Author comment on review comment #2 for manuscript acpd-2005-0336

We are grateful to the referee's overall positive comments and suggestions. Please find below our point-to-point reactions in italic.

This paper presents a comprehensive set of BrO profile measurements with the aim of making these available to the satellite community for the purposes of satellite validation. This is a valuable contribution to the field of remote sensing of BrO and I encourage its publication in ACP. It is a well-written paper, coordinating a large number of groups with different techniques and methodologies. Validation of satellite measurements is extremely valuable and this paper paves the way for BrO.

A major weakness of this work is the inversion technique employed for the SAOZ measurements, with developments now in the field of profile retrievals since the early SAOZ work ie [Hendrick, et al., 2004], and Butz et al. 2005 who are coauthors on this work, this could be improved upon.
This paper focuses on the SCIAMACHY profile retrieval of the Harvard group, with this work in review. The published results of the Bremen SCIAMACHY group should be acknowledged and compared - [Sinnhuber, et al., 2005]. This could be reviewed in the introduction along with other methods of measuring BrO. This balloon data set will provide a useful foundation for validation of other satellite measurements of BrO such as OSIRIS (potential for profiling) and more recently OMI(column). A paragraph in the introduction of the satellite instruments that have/intend to measure BrO from space would add to this paper.
A sentence was added on page 13037, line 17, regarding other satellite measurements: 'The method and results discussed in this study are also of value for the validation of other existing satellite measurements of BrO (e.g. OMI) or satellite instuments that intend to measure BrO in the future (e.g. GOME-2).' We refrain from dicussing details of other satellite measurements, since this would go beyond the scope of the present paper.

The DOAS technique section 2.1.2 is slightly confusing - the DOAS spectral retrievals and then profile inversion technique employed. The DOAS technique section 2.1.2) quotes the three different methodologies 1) direct comparison of SCDs Harder

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(a look up table with no weights??) and 2) Least squares with constraints (which constraints?) Butz and 3) Pundt 2002 using onion peeling.

Page 13020, line 7-10: DOAS is actually only applying methods 1) and 2). 3) is only used by the SAOZ team. Since in this study method 1) is not used, the sentence was replaced with: 'Profile information on stratospheric BrO was obtained by a least-squares profile inversion technique (Maximum A Posteriori) (rodgers2000book).'

A table of the different balloon experiments (ie introduce the names LPMA/DOAS, TRIPLE (in situ), SOAZ (these are the three names used for the rest of the paper)), and their different analyses ie. technique solar occulation, vs limb sampling, spectral window and fitting method ie Aliwell, vs Windoas, cross-sections used. Then the profile inversion technique; least square unconstrained, constrained least squares, or onion peeling. Indicate whether multiple scattering is included, whether the photochemistry is included in the profile inversion (check boxes maybe?). SOAZ neglect both of these effects? A line of the table also could be devoted to the SCIAMACHY retrieval, it would make it clearer to the reader which spectral analyses are used and which profile inversion technique and what approximations are employed etc. Is the direct method of Harder 2000 ever used?

For all instruments, details on the retrieval and the instrumental features have been published before and the individual techniques are well established. The present study does not intend to directly compare the different retrievals and methods of balloon BrO profiling, but to use the established techniques to present a best possible BrO reference set for SCIAMACHY validation. In section 2.1 and 2.2 we tried to make clear the main features of the individual retrievals, including changes and improvements compared to previously published material. We therefore refer the interested reader to the cited material and the references within.

An error is quoted from the Pundt 2002 paper for the effect of neglecting the photo- chemistry and multiple scattering. Figure 2 in this paper (Dorf) displays a photochemical change that is larger than any of the cases shown in figure 5 of Pundt, (ascent finishing at Ÿ90) thus the error of neglecting the photochemical change might be larger for these balloon flights than those quoted in Pundt 2002. Do the authors have an idea how large this error is? Since the inversion of Butz takes photochemistry and multiple scattering during the ascent into account could the SOAZ flights be reanalyzed with this retrieval? Or at least could the Butz retrieval be used to quantify the error in neglecting multiple scattering and photochemistry in the ascents etc? The use of the onion peeling technique would also increase this photochemistry error sensitivity as the subtraction of the amount in the upper levels with less BrO (due to the photochemical conversion) would result in more BrO being assigned to the lower levels? A large portion of this paper is devoted to explaining why photochemistry is important (which it is and is handled in a novel and rigourous way mostly) for satellite validation exercises, yet performing the balloon profile retrievals neglecting photochemistry is slightly contradictory.

General comment: Only photochemistry is taken into account in the Butz et al. inversion, but no multiple scattering.

Specific comment on the SAOZ profile inversion:

The SAOZ profile retrieval makes use of the onion peeling technique in a spherical atmosphere divided in 1 km thick shells in which optical paths are calculated by ray-tracing. In the solar occultation configuration where weighting functions are well defined, all retrieval techniques are equivalent and there is no need for using a priori information. In the case of SAOZ-BrO ascent, since the sampling is higher than 1 km (4 -5 data points / km) and the measurements are relatively noisy compared to other species, slant columns are smoothed with a triangular filter of 3 km half-width. The vertical resolution of retrieved profiles is thus limited to 3 km. The duration of the balloon ascent from the tropopause to 30 km is about 40 minutes, during which the SZA increases at sunset by about 8° at the tropics and 3° at the polar circle. The column photochemical change compared to the time of the balloon passing at 20

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km has been simulated with the photochemical PSCBOX box-model (Hendrick et al. 2004). In the case of the Bauru flight, the coefficient varies from 0.98 at 78° SZA at 14 km to 1.06 at 85.6° at 29 km. In the case of Kiruna it varies from 0.92 at 85.4° SZA at 16 km to 1.01 at 88.1° at 29 km. In both cases, the column change of 6-8% during the balloon ascent is negligible compared to measurements error. The impact of scattered sunlight on SAOZ measurements is described in Pundt et al. (2002). Multiple scattering is negligible at SZA <93-94°. What is important with the SAOZ -5°, +15° elevation and 360° azimuth FOV, is the reflected light in case of clouds and the diffuse single scattered light. Measurements below a cloud layer are easily removed by a colour index method. The contribution of diffuse light compared to direct sunlight falls rapidly above the lower layers of the atmosphere (the sky is dark) and that of reflected light is dropping at large SZA. The total contribution at 350 nm, 20 km, 0.8 albedo and 80° SZA is of the order of 20%, dropping to a few percent at 85-88° at float altitude around 30 km. Model simulations have shown that the impact on BrO mixing ratio does not exceed 0.3-0.4 ppt.

Hendrick, F., B. Barret, M. Van Roozendael, H. Boesch, A. Butz, M. De Mazière, F. Goutail, C. Hermans, J.-C. Lambert, K. Pfeilsticker, and J.-P. Pommereau, Retrieval of nitrogen dioxide stratospheric profiles from ground-based zenith-sky UV-visible observations: validation of the technique through correlative comparisons, Atmospheric Chemistry and Physics, 4, 2091-2106, 2004

Pundt, I. J.-P. Pommereau, M.P. Chipperfield, M. Van Roozendael and F. Goutail, Climatology of the stratospheric BrO vertical distribution by balloon borne UV-Vis spectrometry, J. Geophys. Res., DOI 10.1029/2002JD002230, 2002.

Perhaps of greater concern is the neglecting of multiple scattering in the inversions, especially for assigning the amount of BrO above the float, since multiple scattering acts as an offset in the modelled AMFs. Could a more accurate error be

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assigned, or could the inversion be performed with multiple scattering included using the models of some of the other coauthors ie Hendrick, Butz etc.

For SAOZ: Please see statement above.

For DOAS: Scattering does not play a significant role for the direct sun observations with a narrow field of view. It can in fact be shown that the total contribution of single scattering, even for low altitudes during ascent, is far less than 1%. Multiple scattering is of no relevance for DOAS.

Minor comments:

The order of the affiliations should be matched with the order of the authors.

Done.

Quoting unpublished results of Dorf on line 20 page 13014 is not necessary as published results are available for this number.

Quoting unpublished results of Dorf et al. is necessary since not all the information to make such a statement is given in the other published results.

Page 13014 line 22: 60-70% for BrO/Bry of Lary (1996) seems high - check by comparing with SLIMCAT used in this work, and [Avallone and Toohey, 2001] of 0.45-0.6 for the measured partitioning of bromine.

Comparison with SLIMCAT confirms the 60-70%

Pg 13016 line 2: sentence beginning 'However, the large diurnal variation... even if a perfect match of both observations ie. In one altitude range at one time.' If you could measure at the same time, you would measure at the same SZA, the problem perhaps is the duration of different observations? The balloons taking Ÿ1hour 20 and

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the satellite being a much shorter measurement.

We agree that this sentence is slightly confusing. It was changed to: 'However, the large diurnal variation of the BrO radical, the different measurement geometries, the duration of the different profile observations and, to a lesser extent, presumably small spatial gradients in total stratospheric bromine (and thus BrO) prevent a direct comparison of the balloon-borne and satellite limb measurements.'

Page 13024 line 19: and 22 mismatch should be match??
We really mean a mismatch in order to emphasise the possible time delay (of one hour or less) and the differing location (of less than 500 km)

Figure 3 has no backward match with SCIAMACHY - remove panels? Is this the only Triple match?
Unfortunately no 'backward' Triple match is available. We would like to keep the upper panel of the plot since it shows the results of the photochemical scaling, and therefore the validation profile.

Page 13033 line 10 fo should be for
Done.

Avallone, L. M., and D. W. Toohey (2001), Tests of halogen photochemistry using in situ measurements of ClO and BrO in the lower polar stratosphere, Journal of Geophysical Research Atmospheres, 106, 10411-10421.

Hendrick, F., B. Barret, M. Van Roozendael, H. Boesch, A. Butz, M. De Maziere, F. Goutail, C. Hermans, J. C. Lambert, K. Pfeilsticker, and J. P. Pommereau (2004), Retrieval of nitrogen dioxide stratospheric profiles from ground-based zenith-sky UV-visible observations: validation of the technique through correlative comparisons,

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Atmospheric Chemistry and Physics, 4, 2091-2106.

Sinnhuber, B. M., A. Rozanov, N. Sheode, O. T. Afe, A. Richter, M. Sinnhuber, F. Wittrock, J. P. Burrows, G. P. Stiller, T. von Clarmann, and A. Linden (2005), Global observations of stratospheric bromine monoxide from SCIAMACHY, Geophysical Research Letters, 32.

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