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Atmospheric Measurement Techniques Discussions

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Interactive Comment

Interactive comment on "New retrieval of BrO from SCIAMACHY limb: an estimate of the stratospheric bromine loading during April 2008" *by* J. P. Parrella et al.

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The authors would like to thank Reviewer 1 for the helpful comments on our paper. We have revised segments of our paper to reflect these comments, and address each suggestion and question in this document below.

The reviewer's comments and questions are copied here in quotations. Our responses are written below each comment block quotation.

Reviewer 1: "What has been improved on the technical side to reduce uncertainties in the new retrieval compared to existing BrO profiles from other groups? (...) In order to

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really help reducing existing uncertainties the manuscript should provide more details of how and why the current approach leads to improved BrO retrievals. Ideally I would like to understand why there are differences to previous BrO retrievals, see how the new retrieval compares to existing retrievals and how the comparison with the balloon-borne observations improves for the new retrieval. This would require some major revisions to the current manuscript. However, I believe such an effort is worthwhile and I would like to encourage the authors to consider submitting a revised manuscript."

Response: We interpret the above comments on our paper as questioning 2 things in particular: (1) How is our BrO retrieval method different? (2) Why is this approach better, or why did we select this among other documented approaches. We note that Reviewer 2 was satisfied with the methods we chose for this paper.

We clarify the first question here. The primary difference in our method is in the spectral fitting step, where slant column amounts of BrO (#/cm2) are fitted from the calibrated SCIAMACHY radiance data and the complete cross-sections. The other groups retrieving BrO from limb geometry near-UV measurements apply a linearized version of the DOAS algorithm. In their process, they first fit 3rd or 4th order polynomials to the observations (mostly to adjust for atmospheric scattering) and the laboratory cross-sections, which are then used as basis functions, and subtract the respective polynomials from the data and cross-sections. After this pre-processing, they then fit the spectra. In our method, we fit one set of polynomial parameters at the same time as fitting BrO and other trace gas slant column depths from the data – this requires a non-linear least squares approach, which is what we implemented.

Our response to the second question, why we chose a non-linear fitting algorithm, builds the explanation in the previous paragraph. It is considered best practice in scientific computing to fit all parameters at once, in the same step (e.g. C. Molar, 2004). This is what our algorithm does, and is a difference between our work and the previous literature (Sinnhuber et al., 2005; Sioris et al., 2006; McLinden et al., 2010; Rozanov et al. 2011). Fitting different parameters in series can result in unwanted biases and

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problems with error propagation (Carlotti, 1988). We agree with Reviewer 1 that a detailed comparison between a full non-linear least squares approach and the linearized approach would be very interesting and useful, and encourage that as the topic for future research. However, we believe that this is outside the scope of our current paper, and is not necessary to justify the use of well-established algorithms such as those that we use in our current paper (Lïndstrom and Wedin, 1988; Caspar and Chance, 1997; Chance, 1998; Chance et al., 2005)

Reviewer 1: "How do the new BrO retrievals compare to existing BrO retrievals from SCIAMACHY? How does the new estimate of stratospheric bromine presented here provide new constraints?"

Response: As we described above, our non-linear least squares approach to the spectral fitting step is new compared to previous limb BrO retrievals. Our estimated range of Bry mixing ratios in Figure 6, as our plots of estimated Bry in Figure 8 provide an implicit comparison to existing retrievals of SCIAMACHY BrO. We agree with Reviewer 1 that detailed intercomparisons between retrieval algorithms is very useful, and that more profile comparisons between our retrievals and those in existing retrieval algorithms should be the topic of future research. However, we believe that introducing a new algorithm for BrO limb retrievals from SCIAMACHY is a worthy paper on its own, and do not think that a detailed intercomparison is necessary for introducing our new retrieval algorithm.

Reviewer 1:"Specific Questions: 1. More information should be given regarding the comparison with the balloon observations. How/why were the five balloon profiles selected for comparison?"

Response: These observations were provided to the authors with photochemical corrections already applied to match the conditions of the shown SCIAMACHY retrievals. Since they were easily accessible to the authors, we chose these balloon observations as a test to demonstrate that our profile retrievals are within the range of a set of

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previously documented, and independent observations.

Reviewer 1:“Are these observations in 2003 and 2004 all the available balloon-borne BrO profiles? Please provide more details on the BrO retrieval from the balloon observations to give the reader an idea of whether these observations can be regarded as fully independent, or, e.g., if they both depend on the same spectroscopy, i.e. uncertainties in the cross sections cancel out to first order.”

Response: The retrieval methods used for the balloon-borne observations are documented by Dorf et al., (2006). They use linear DOAS spectral fitting and an “onion-peeling” approach (layer-by-layer inversion) to for the inversion of slant column densities of BrO to mixing ratio. This is independent from our approach, which uses non-linear least squares fitting for BrO SCD estimates followed by optimal estimation of all layers in the atmosphere simultaneously. Additionally, they do not use the Wilmouth et al. (1999) cross sections for BrO that we use in our work. We cite the paper with their methods, and decided not to go into detail on the Dorf et al. (2006) approach. Additionally, their observations come from a different instrument, and thus have different spectral resolution and instrumental artifacts, requiring different calibration steps and different decisions during to fit the data. For these reasons, the fitting methods of Dorf et al. are sufficiently independent from our own methods shown in this paper. The authors believe that a citation of Dorf et al. (2006) is sufficient to document the methods used by Dorf et al. in their work on BrO balloon observations. We also note that this seems to be the opinion of Reviewer 2.

Reviewer 1:“Little is said about the photochemical correction here.ï£ij How critical is this? How different are the local times of observations?”

Response: The photochemical corrections are also described in Dorf et al. (2006), including an analysis of errors. These observations, with the exact same photochemical correction, have additionally been used in cross-validation for several existing retrievals of BrO mixing ratio from SCIAMACHY limb measurements (Rozanov et al., 2011). For these two reasons, the authors believe these balloon observations, with photochemical interpolation between solar times, to be very well suited for a comparison with our SCIAMACHY retrievals. The authors decided not to go into detail on the methods of Dorf et al. (2006) since it has already been used for intercomparison in Rozanov et al. (2011), and is documented in these two previous papers.

Reviewer 1:"It is stated (p.8034) that uncertainties in the photochemical modeling dominate the uncertainty in inferred total bromine. Does this represent also a major uncertainty here in the comparison with the balloon observations? In any case, the comparison should be made more quantitative. How does this study compare to the numbers given by Rozanov et al. (2011)?"

Response: We interpret this question to be asking whether the uncertainties in Bry photochemistry could significantly affect the diurnal cycle of BrO mixing ratios. The interpolation made on the balloon observations, along with errors incurred, is described by Dorf et al. (2006) and Rozanov et al. (2011). The authors do not consider that further discussion of these methods will add to our current paper, as the balloon measurements have been well-vetted. The Rozanov et al. (2011) SCIAMACHY BrO retrievals compare well with these same balloon observations as well.

Also, the authors do not agree with the Reviewer's comment, "the comparison should be made more quantitative." We would like to note that Reviewer 2 does not believe there is any more quantitative work to do for these 5 comparisons either.

Reviewer 1:"2. The BrO/Bry ratio is largely controlled by O3 and NO2 (p.8033, l.28) and uncertainties in O3 and NO2 may contribute to the overall uncertainty in modeled BrO/Bry. It is stated (p.8032, l.1) that for the BrO retrieval O3 and NO2 are simultaneously fitted. Have these O3 and NO2 results (or O3 and NO2 from any other SCIAMACHY retrievals) been used to constrain, or at least test, the O3 and NO2 in the photochemical calculation of BrO/Bry?"

Response: No, they have not. However, the BrO/Bry ratios from Salawitch et al. (2010)

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were generated specifically for April 2008 (the same time period we look at in our paper), and used assimilated fields for O3, NO2, as well as detailed meteorology, that rely on multiple observations. This is documented in their paper. Comparisons of our O3 and NO2 slant columns is outside the scope of this paper.

Reviewer 1: “The final sentence (p. 8035, l. 23) states that uncertainties in chemical kinetics dominate the uncertainty in inferred bromine loading, which is a strong statement! Does this include uncertainties in the concentration of O3 and NO2?”

Response This does not include uncertainties in the concentration of O3 and NO2. Uncertainties in the chemical kinetics drive uncertainty in the conversion between BrO to Bry. This is shown and discussed by Salawitch et al. (2010) – we simply apply the Salawitch error estimates to our final errors. We also note that our final error estimates are similar to those given by McLinden et al. (2010) and Hendrick et al. (2008), which both used different instruments and different retrieval methods. For these reasons, stating the chemical kinetics as the primary driver for our uncertainty in Bry estimates is not controversial.

Reviewer 1:“3. It is stated that "including cross sections for O2-O2, OClO and additional temperatures for ozone ... occasionally caused strong correlations with BrO" (p.8025/8026). Shouldn’t that worry one that there are spectral interferences with these other absorbers and how was this accounted for?”

Response: The authors thank Reviewer 1 for bringing this up. We forgot to mention that the concentrations of O2O2 and OClO are within spectral noise for the range of measurements considered in our work. We have changed this sentence in the text to make note of this.

Reviewer 1:“4. SCIAMACHY/ENVISAT (p.8020): I suggest including information for which time period SCIAMACHY provided data and to specify a bit clearer which different BrO profile products already exist. All relevant papers are given, as far as I can tell, but it is not spelled out so clearly which studies have provided BrO from SCIAMACHY

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observations."

Response: Our work focused on April 2008, when we had detailed BrO/Bry information from Salawitch et al. (2010), as we have stated in the original document, because this is the period of the NASA ARCTAS Campaign. SCIAMACHY operated between 2002 and 2012.

Reviewer 1:"p.8018, l.26: remove "and to the stratosphere" "

Response: This has been modified in our final version.

Reviewer 1:"p.8019, l.5: the reference to Brinckmann et al. (2012) seems out of place here as this study did not deal with limb geometry satellite observations."

Response: Thank you for catching this error, we have now corrected it.

Reviewer 1:"p.8032, l.10: 'these errors are dominated' or 'these are errors dominated'"

Response: "these errors are dominated" is correct – we were referring to "Systematic model errors" when writing "these errors".

Reviewer 1:"p.8034, l. 11: What is DOFS? Plural of DOF (degrees of freedom)? This is currently introduced in the caption of Fig.2 and should be introduced also in the text."

Response: Thank you for catching this mistake. DOFS is intended to be Degrees Of Freedom in the Signal (DOFS), not the plural of DOF. They are often used interchangeably in the literature, and picking one or the other may be semantics. We have included a parenthetical with "Degrees of Freedom" written in the final version.

Reviewer 1:"Fig. 6: What causes the low values of BrO at high northern latitudes for April 10-12? Is this a plotting artifact?"

Response: There is more variability in data at higher latitudes because the signal-to-noise in the spectral observations is greater than at lower latitudes – due to the weaker light intensity reaching the detectors. Also, more data is thrown away in our algorithm for these high latitude cases, because the spectral fitting routine often fits SCDs of BrO that are smaller than the fitted standard deviation.

Reviewer 1:“Fig. 8: It is regrettable that calculated BrO/Bry ratios are available only for the northern hemisphere. In this case the figure range can be restricted to northern hemisphere only.”

Response: If possible, the authors would like to show all data that we have produced for this work and leave the figure as is.

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