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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Retrieval of stratospheric and tropospheric BrO profiles and columns using ground-based zenith-sky DOAS observations at Harestua, 60° N" *by* F. Hendrick et al.

Anonymous Referee #2

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General comments

The manuscript entitled "Retrieval of stratospheric and tropospheric BrO profiles and columns using ground-based zenith-sky DOAS observations at Harestua, 60° N" presents a retrieval algorithm for atmospheric BrO profiles based on zenith-sky DOAS measurements of the BrO SCD. The algorithm is applied to data obtained from an instrument based in Harestua, Norway. The resulting profiles and partial tropospheric and stratospheric columns are compared to collocated measurements from balloon and satellite as well as to results from chemical models.

The paper is well structured and scientifically sound. In particular, the validation of the

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algorithm and the good agreement between ground-based, satellite and balloon borne measurements as well as model results presented in the manuscript provides further evidence that DOAS measurements of scattered sunlight are well suited for the retrieval of profile information of atmospheric trace gases. Apart from forward model parameter errors, which should be discussed in more detail, a comprehensive discussion of the errors of the retrieved quantities is presented.

The paper provides further evidence of a tropospheric BrO background, which is expected to have a significant impact on the photochemistry in the free troposphere (e.g., von Glasow et al., 2004). I recommend the publication of the manuscript after some minor modifications as described below.

Specific comments

Page 8865, Line 14: It is stated that 'The ground-based DOAS ... technique has been extensively used to study and monitor ozone depletion ... in the troposphere ...'. However, scattered light DOAS instruments are usually not capable to measure tropospheric ozone.

Section 2: One single reference spectrum for a whole year has been used for the BrO profile retrieval. As the authors mention, this significantly increases the information content of the measurements. The RSCD is determined in a two-step method using the Langley-Plot method. This method is somewhat circular: Assumptions need to be made on the BrO profile shape (which is particularly uncertain in the troposphere) to calculate AMFs which yield VCDs and the RSCD. This RSCD is in turn used as a model parameter to determine BrO profiles. Quantities which are not directly measured should actually be retrieved, and a retrieval of the RSCD by the optimal estimation algorithm would automatically ensure the calculation of 'correct' AMFs in a sense that the BrO profiles serving as input for the radiative transfer model are identical with the retrieved profiles. The authors have spent a lot of effort in determining the fraction of tropospheric BrO, although this is actually determined by the profile retrieval algo-

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rithm. It is necessary to check the consistency between the initial assumptions and the retrieval results. How does this factor f, and also the BrO profile used for the AMF calculation, compare with the results from the retrieval? How does the Langley plot look like if the profile from the retrieval is used as input for the AMF calculations?

A tropospheric BrO fraction of 30% is chosen, although Fig. 1 suggests that 40% is the better choice. Why? What is the average fraction obtained by the retrieval?

Table 1 lists the RSCDs determined by the Langley-plot method and the corresponding standard deviations. Provide an estimate for the systematic errors of the RSCDs, e.g. due to uncertainties in the AMF calculations (uncertainties of the RTM, BrO profile shape, aerosols and other RTM parameters).

Section 3: On which altitude levels are the BrO profiles retrieved?

Page 8672, last paragraph: not the statistical measurement errors, but their square (i.e., the variance) must serve as diagonal elements of S_ε.

It is stated that the fit error is dominated by random detector noise. If this would be the case, one would expect that the magnitude of the scattering of the data is in the order of the size of the error bars. However, the diurnal variation of the BrO SCD shown in Fig. 6 seems to be much smoother than one would expect from the error bars, indicating that there are systematic errors (i.e., systematic residual structures). It would be useful for the reader to see a typical example of a BrO fit.

It is stated that the a priori covariance matrix is 'generally not known'. This is not true, and if the optimal estimation method is applied strictly, the a priori covariance matrix needs to be constructed from other measurements or models, for example on the basis of a climatology (this is discussed in detail in the book of Rodgers). For the stratospheric BrO profile, one could for example construct a covariance matrix from a climatology based on the output of the RTM.

Section 4.2: Here, the error budget of the retrieved BrO profiles is discussed. In this

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context, it is confusing to see the measurement error as an error component of the profile. Instead, the measurement error propagates into the retrieval error as the so-called retrieval noise, $S_m = G_y S_\varepsilon G_y^T$, with G_y being the gain matrix. Do the values referred to as 'measurement error' in Table 2 really refer to the measurement error or to the retrieval noise?

As already discussed by Referee 1, a thorough discussion of the model parameter error is missing. A value of 12% is assumed, but it is not detailed where this number comes from and whether any sensitivity tests have been performed to estimate the individual error components. I think it is not sufficient to adapt a model parameter error from the Schofield paper, which deals with a different measurement technique (including direct sunlight) and uses different forward models. Apart from the uncertainties in the chemistry model and the error in RSCD, these could be uncertainties in the RTM, both since the DISORT algorithm is only an approximation and due to errors in the RTM parameters. In particular, what kind of aerosol scenario has been used, and how do the uncertainties in aerosol loading propagate into the retrieved profiles, both regarding the retrieval itself and the determination of the RSCDs?

Section 5, last paragraph: It is first mentioned that GB measurements underestimate the BrO total columns, and then that SCIAMACHY underestimates the other measurements. Since it is not clear which of the measurements provides the 'true' values, one should instead state that the measurements disagree.

Section 6: The tropospheric BrO column is slightly larger at am than at pm. As a priori for the diurnal variability in the troposphere, the chemistry of the lowermost stratosphere is used. Since stratospheric BrO concentrations are slightly higher in the morning (e.g., Sinnhuber et al., 2002), the question arises to what extend this effect is caused by the a priori.

Section 7: The authors argue that the higher measured tropospheric BrO values in spring compared to the TOMCAT model might be caused by bromine-rich air from

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the Arctic. Since mid-latitude transport of polar air with high bromine content is a very interesting issue, it would be very nice to see whether this can be confirmed by trajectory models.

Technical corrections

Abstract, 2nd sentence: Replace with 'The sensitivity of the zenith-sky observations to tropospheric BrO is increased by using a constant reference spectrum observed during clear-sky noon summer conditions for the spectral analysis'.

P 8664, L20: replace 'feature' with 'finding'. The whole sentence is not clear and requires re-structuring.

P 8664, L23: Delete 'corresponding'.

P 8664, L25: Replace 'used' with 'used in this study'.

P 8664, L26: Replace '+ 6 extra pptv' with 'and additional 6 pptv'.

P 8665, L14: Move 'to the stratosphere' before 'would have a...'.

P 8667, L25: Define the acronym IASB-BIRA.

P 8668, L17: Replace 'different' with 'fixed' or 'single'.

P 8671, L24: Replace 'for the' with 'of'.

P 8672, L8: Replace 'of unique solution to the' with 'of a unique solution of the'.

P 8673, L7: Replace 'key parameter in' with 'key parameter for'.

P 8674, L6: Replace 'limited' with 'small'.

P 8675, L5:: Use 'T' instead of 't' for the transpose, as in Eq. 4. The transpose requires no explanation in the text.

P 8677, L3: Replace 'Optimal Estimation Method' with 'OEM'.

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P 8677, L26: Replace ’kernels’ with ’kernel’.

P 8678, L9: Replace ’is observed’ with ’occurs’.

P 8679, L2, 26, 29: Replace ’equator’ with ’Equator’.

P 8679, L20: Replace ’raison’ with reason’.

P 8679, L27: Replace ’off-set’ with ’offset’.

Section 6, 1st sentence: Add ’as described in Section 4.2’ to the sentence.

Section 7: As for the TOMCAT model runs, which are labelled with ’run 1’ and ’run 2’, also the SLIMCAT model runs should be labelled with ’run 1’ (standard run), ’run 2’ (6 ppt offset) and ’run 3’ (8 ppt offset).

Figure 10: Please add error bars to the tropospheric and stratospheric VCDs retrieved from DOAS measurements.

References: Put all publication titles in lower case.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 8663, 2007.