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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A global stratospheric bromine monoxide climatology based on the BASCOE chemical transport model" *by* N. Theys et al.

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Nicolas Theys: Author response to Referee #2

Review of 'A global stratospheric bromine monoxide climatology based on the BASCOE chemical transport model' by Theys et al. submitted to ACP.

This paper presents a new approach to design a global climatology of stratospheric BrO specifically intended for the retrieval of tropospheric BrO columns from the satellite nadir instruments but will also be available for other applications. The parameterization used for this climatology is evaluated in the manuscript based on 3 years of output from the BASCOE chemical transport model. The dynamics impacting on the stratospheric BrO distribution is accounted for by a parameterization based on ozone columns, the

effect of photochemistry on stratospheric BrO is determined by using NO2 columns and the SZA. Both ozone and NO2 outputs from BASCOE used to build the BrO climatology have been extensively validated within this paper as well as in other previous publications and the authors found that the important changes in stratospheric ozone and NO2 are consistently reproduced by the model (except for SH ozone hole conditions). They conclude that the overall uncertainty on stratospheric BrO is lower than 30%.

Reply: We are grateful to the referee's comments and suggestions.

General comments:

1) I find the research described in this publication to be of great interest but it leaves me rather confused re the actual purpose of this publication: The authors go through quite some trouble to show that BASCOE can model observed BrO well (section 3.2, also: 'the model simulations have been optimized for bromine chemistry and budget')

Reply: Realistic model settings for the bromine sources and chemistry have been implemented, which are fully consistent with our current understanding of stratospheric bromine as summarized e.g. in the last WMO report (2007, Scientific assessment of ozone depletion). In that respect, and considering that no assimilation of active bromine measurements has been performed, we believe that the consistency between the simulated BrO profiles and the different sources of correlative data is highly satisfactory.

but then they don't use the actual BASCOE BrO output to build up the climatology. Instead they build the climatology based on ozone and NO2 output- this is very well motivated and convincing as a method but why not use straight the BrO output from BASCOE - would that no make more sense? This really also begs the questions how would this 'straight out of the model' BrO climatology compare to the BrO that is 'built' based on NO2 and ozone? If the actual climatology is the important message here, then why not use the BASCOE BrO fields straight? Or is a simplified parameterization

ACPD

8, S9714–S9719, 2008

Interactive Comment

based on ozone and NO2 the main message which can also be used if no straight BrO output would be available but then the validation of BASCOE BrO is a little confusing (good to see though that is works ok). I think it really would be important to get this message straightened out - or please do tell me what I am missing here, I am having a bit of a head cold at the moment, so hope that this is not impacting on my understanding.

Reply: The key idea of the proposed BrO parametrisation is that it is achieved through two distinct climatologies: one for total inorganic Bry and a second one for the activation ratio BrO/Bry. Both climatologies are, of course, built using the BASCOE 3-D chemical fields of BrO and the other inorganic bromine species included in the model. As explained in sect. 4, the climatologies rely on a tabulation of the modeled Bry and BrO/Bry profiles respectively for different O3 and NO2 columns, calculated from the corresponding BASCOE modeled O3 and NO2 profiles. It is shown that this simple classification using ozone and NO2 columns is able to reproduce the original modeled stratospheric BrO profiles with a precision better than 14%. On the other hand, the BASCOE O3, NO2 and BrO output (used to build up the climatologies) have been consolidated independently, through comparisons using an extensive set of observations. The parameterization of stratospheric BrO as presented in this study, has several advantages for its primary application, i.e. the retrieval of tropospheric BrO columns from satellite nadir observations:

- This approach can be used even if no model data is available.

- The O3 and NO2 columns simultaneously measured by the satellite instrument can be used as a constraint for the estimation of stratospheric BrO profiles. This approach is actually more representative of the sounded pixels than a straight use of the (low resolution) model fields.

- The simplicity of the method.

The authors went through the manuscript and have clarified the abstract and the intro-

ACPD

8, S9714–S9719, 2008

Interactive Comment

duction, so that the reader understands better the key concept of this study.

2) No doubt such a global stratospheric BrO climatology is a very useful tool but it certainly has to be applied with a lot of care specially when used to retrieve tropospheric BrO from satellite observations. The authors discuss briefly e.g. in section 4.1 that 'misrepresentation of important patterns of stratospheric BrO will lead to artifacts on the retrieved tropospheric BrO columns'. So, although this will be quite a powerful method I strongly believe that a thorough validation of the resulting tropospheric BrO columns by either balloon observations, ground-based retrievals or in-situ observations will be a very important reality check.

Reply: This is precisely the reason why the present climatology is designed to be able to reproduce the important variations of stratospheric BrO by using dynamical and chemical indicators (in practice, they are measured quantities, so it guarantees that the sounded air masses are optimally represented). Furthermore, efforts have been devoted to the validation of the BASCOE BrO output in sect. 3.2. However, we agree with the referee that the retrieval of tropospheric BrO from satellite observations and the present climatology requires great care to achieve good accuracy. We are also convinced that the derived tropospheric BrO results will have to be confronted to external measured data in order to test and validate the method. Though this is beyond the scope of this paper, we agree it has to be emphasized in the text, as well as a discussion of the error on the retrieved tropospheric BrO columns (see below).

3) Given that the overall uncertainty in this BrO climatology could be as large as 30%, will this be of sufficient quality so that the climatology can be used for its main design purpose, i.e. to pull out tropospheric BrO from satellite columns? I do understand that this will be discussed in detail in a future paper (as stated in the conclusions) but since this is also the main motivation for this paper, it would be helpful if the authors could elaborate on this a little.

Reply: The error on the stratospheric BrO columns evaluated by the BASCOE climatol-

ACPD

8, S9714–S9719, 2008

ogy is limited by the accuracy of the modeled BrO, and only slightly by the error linked to the parameterization presented here (see section 4.5). We estimate that an error on the retrieved tropospheric BrO columns of about 25% for bromine explosion events and of 50% for free-tropospheric BrO background conditions may be attributed to the stratospheric BrO correction. The precise value depends on the observation geometry, surface reflectivity and the BrO partitioning in both stratosphere and troposphere. However, we believe that valuable results related to the spatial and temporal variations of tropospheric BrO can be obtained from the combined use of satellite BrO observations and the present climatology. Moreover, this error will likely decrease in the future as consolidated kinetic and photochemical data, and improved measurements of inorganic and organic bromine compounds will be available.

4) Another minor comment: Under section 3.2.1 the authors provide some background on the BrO measurements used for the comparison. Stratospheric BrO profiles retrieved at 80 SZA are used to compare with the model e.g. in Fig 4. That is all looking good but they then say that the BrO profiles can be converted to any SZA by using a photochemical model. Certainly also true, but that then would not work anymore as a measurement to model (BASCOE) comparison but a photochemical box model to BASCOE comparison, right? Do I understand this right that there is only one independent value, e.g. for 80 SZA retrieved from the observations? I just felt it could potentially sound a bit misleading as in the text.

Reply: We agree with the referee that there is some misunderstanding on this part. The ground-based BrO profiles are retrieved from twilight observations (70-92°SZA). Therefore they are representative of a SZA of about 80° without any photochemical corrections. Then they are compared to the BASCOE BrO profiles evaluated at this particular solar zenith angle (and at the location of the observation). For this, only an interpolation of the BASCOE BrO fields (model time step: 30 minutes), is used. The photochemical box model is not used for any SZA conversion. In the text, the following sentence in sect. 3.2.1: 'However, the retrieved BrO profiles can be converted to any

ACPD

8, S9714–S9719, 2008

SZA by using the photochemical model' brings some confusion and has thus been removed.

Specific comments:

Page 17593: Hendrick et al., 2008b paper cited only under preparation, might have to be removed if not submitted in time. Reply: The paper of Hendrick et al., 2008b has now been accepted for publication in Atmos. Meas. Techn. Discuss. and will be available on the AMT Discussion website soon. Therefore, the reference has been changed.

Figure 8: Suggest to swap the 2 top plots so the 30-40N is displayed above the 30-40S plot (or swap the 2 bottom plots). Reply: this has been changed.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 17581, 2008.