Global stratospheric BrO based on the BASCOE 3D chemical transport model

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Introduction

Inorganic bromine (Br_y) plays an important role on the atmospheric chemistry (in particular on the ozone budget) in both stratosphere and troposphere. Several organic substances, emitted in the troposphere, are sources of Br_y in the UT/LS region: CH_3Br, man-made halons and a variety of very short-lived species (VSLS), such as $CHBr_3$ and CH_2Br_2 which are mostly of oceanic (i.e. natural) origin. In the polar boundary layer, inorganic bromine plays a major role in spring during complete ozone depletion events, when bromine monoxide (BrO) is massively emitted ("bromine explosion" phenomenon) thanks to autocatalytic heterogeneous photochemical processes. BrO is the most abundant bromine bearing inorganic trace gas during daylight. It is also the only one that is measured routinely. Satellite UV-visible measurements provide the unique opportunity to study the global spatial distribution as well as long term evolution of BrO in both stratosphere and troposphere, and might help to identify the effects of international restrictions (Montreal Protocol and amendments) on the use of the ozone-depleting brominated substances. The evolution of the emissions of ocean VSLS (Salawitch, 2006) and polar spring BrO in a warmer climate are also pending questions. In the framework of the AT-2 and O_3 SAF BrO VS projects, a particular attention has been paid to the development of a new climatology of stratospheric BrO profiles (and columns), with the aim to apply it to the retrieval of tropospheric BrO columns from satellite UV-visible nadir instruments (such as GOME/ERS-2, SCIAMACHY/ENVISAT and GOME-2/MetOp-1). More precisely, the climatology is planned to be used in a residual algorithm (Theys et al., 2004), to correct for the BrO absorption in the stratosphere. The impact of the atmospheric dynamic on the stratospheric BrO distribution is treated by means of Br_y/ozone correlations build from 3D-CTM model results (BASCOE model), while photochemical effects are taken into account using stratospheric NO_2 columns as an indicator of the BrO/Br_y ratio. Details can be found in Theys et al. (2007a).The climatology is intended to be applied for operational retrieval of BrO columns from GOME-2 (MetOp-1) observations within the UPAS environment at DLR. Here, we assess the reliability of the BASCOE reference model results by means of validation using ground-based, balloon and satellite observations.

The BASCOE model

The Belgian Assimilation System of Chemical Observations from ENVISAT (BASCOE, see bascoe.oma.be) is a 4D-Var assimilation system designed for the analysis and forecast of stratospheric ozone and chemical fields.

- horizontal: 3.75° x 5°; vertical: 37 pressure levels, surface→ 0.1 hPa
- 57 chemical species, 200 reactions
- 4 types of stratospheric PSC particles. Surface area density of PSCs and the loss of HNO_3 and H_2O by sedimentation are calculated using a parameterization (no full microphysics scheme).
- sulfate aerosols are implemented in the model using climatological data based on SAGE II (for tropical and mid-latitudes) and POAM III (for high-latitudes) observations.
- driven by ECMWF operational forecasts of winds and temperatures.
- data used: free model run from 1 May 2003→ 30 April 2004; initialized with the SLIMCAT chemical model

Bromine set-ups

- CH_3Br (8 pptv), Ha-1211 (4.6 pptv), Ha-1301 (3.3 pptv), $CHBr_3$ (2 pptv → 6 pptv of Br_y from VSLS ~ at tropopause) => stratospheric Br_y budget ≈ 22 pptv
- Up-to-date bromine reaction rates (JPL 2006)
- The reaction $BrONO_2+O(^3P) \rightarrow BrO+NO_3$ is not (yet) included in the model.

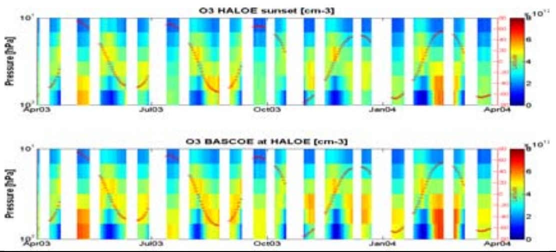
Comparison results

Preliminary (!)

O_3

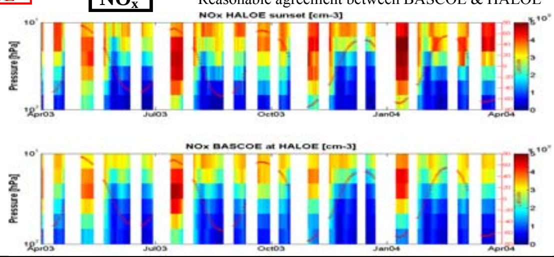
Differences< 10%

BASCOE vs HALOE



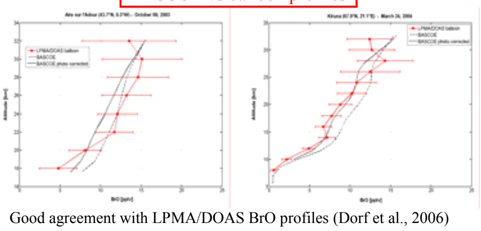
NO_x

Reasonable agreement between BASCOE & HALOE



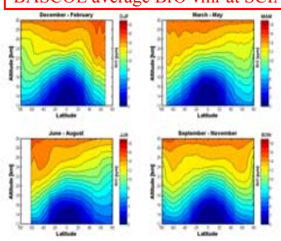
BrO

BASCOE vs balloon profiles



Good agreement with LPMA/DOAS BrO profiles (Dorf et al., 2006)

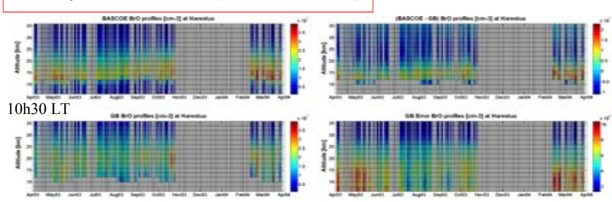
BASCOE average BrO vmr at SCIAMACHY overpass



The distribution of stratospheric BrO is generally consistent (± 1-2 pptv) with SCIAMACHY limb BrO profiles (Rozanov et al.), except at south pole (lower values - under investigation).

BASCOE vs ground-based observations

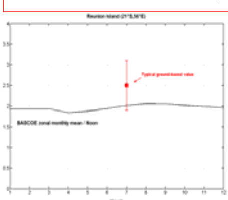
Harestua (60.2°N, 10.8°E): profiles retrieved from zenith-sky DOAS observations (Hendrick et al., 2007)



10h30 LT

The BASCOE profiles lie within the uncertainty of the GB profiles, with a small tendency to overestimate the lower part (<20 km) of the profile and underestimate the upper part (>20 km)

Reunion Island (20.9°S, 55.5°E): columns retrieved from multi-axis DOAS observations (Theys et al., 2007b)



The BASCOE columns are slightly lower than the ground-based estimates, possibly due to:
- uncertainties on the contribution of the VSLS to the BrO column.
- the absence of the reaction $BrONO_2+O(^3P) \rightarrow BrO+NO_3$ in the model.

FUTURE WORK

- Refine the stratospheric aerosol settings (influencing the NO_x/NO_y partitioning and thus the BrO/Br_y ratio).
- Investigate the impact on the results of different scenarios for the VSLS.
- Include the reaction $BrONO_2+O(^3P) \rightarrow BrO+NO_3$
- Extend the comparisons with GB obs. to mid-latitudinal measurements (Observatoire de Haute-Provence: 44°N)
- Rigorous comparisons with SCIAMACHY limb BrO profiles

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