Simulation of BrO Diurnal Variation and BrO Slant Columns : Intercomparison Exercise between Three Model Packages

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Abstract

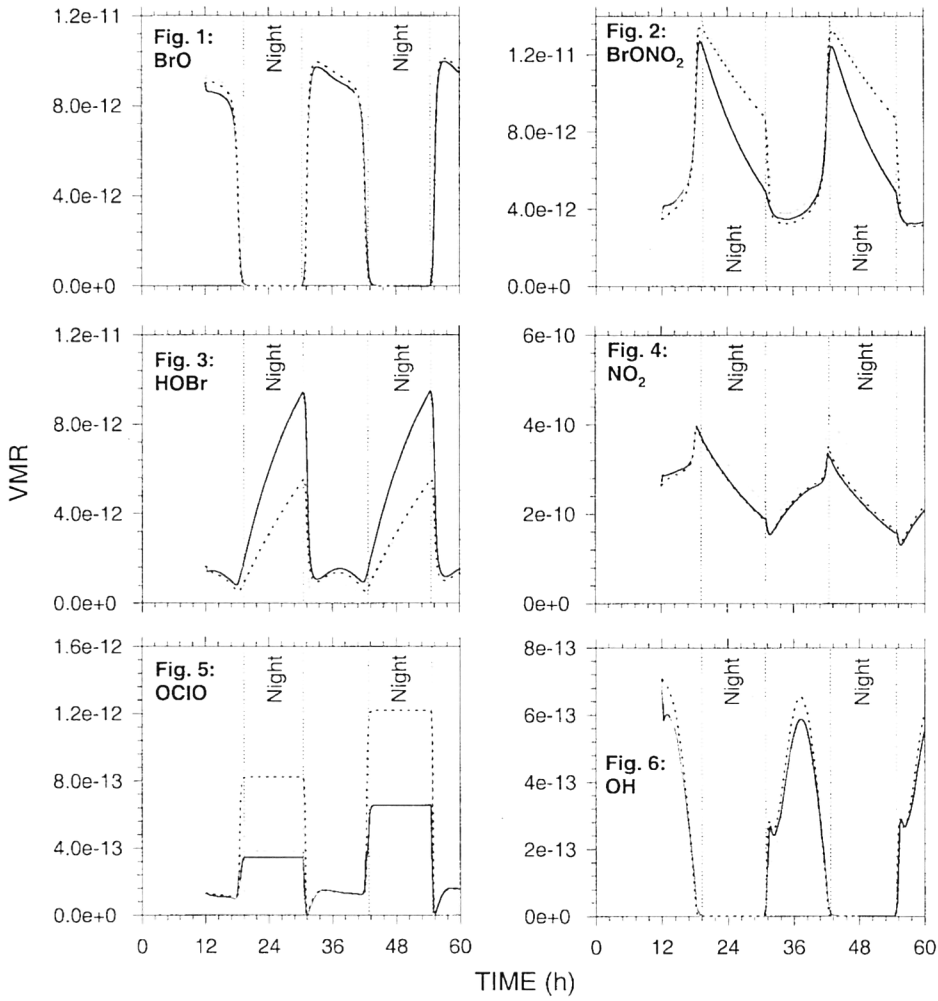
The DOAS-technique (Differential Optical Absorption Spectroscopy) has been extensively used for the monitoring of stratospheric trace species from the ground, balloons and satellite instruments. DOAS observations yield slant column densities (SCD's) as a direct product. In the case of short lived species such as BrO and OClO, the interpretation of the measurements is complicated by the fast diurnal variation of these radicals and by the slant path geometry. Model packages based on 1D photochemical models coupled to ray tracing radiative transfer models enable the simulation with full diurnal cycle of BrO or OClO SCD's. A method to interpret the measurements is therefore to compare them directly to the model calculations.
This paper presents the preliminary results of an intercomparison exercise between three model packages: SLACO (IUP-Bremen), PSCBOX (IASB-BIRA) and SLIMCAT1D (University of Leeds). The intercomparison consists in verifying the consistency between the model packages through tests using common initialisation data and in comparing simulated BrO SCD's to ground-based DOAS measurements at Bremen (53°N) and Ny-Ålesund (79°N).

1. Description of the model packages

- *SLACO (IUP- Bremen)*: the stacked 1D photochemical box-model BRAPHO [1] coupled to a ray tracing model [2]. BRAPHO is based on the ASAD integration routine [3], the heterogeneous chemistry model MPI HET [4], the JPL97 chemistry [5] and the photolysis model PHOTOGT [6]. The ray tracing model (single scattering) calculates the rays in a full spherical geometry using the appropriate local concentrations.
- *PSCBOX (IASB-BIRA)*: the stacked 1D photochemical box-model PSCBOX coupled to a ray tracing model. PSCBOX [7] is based on a Rosenbrock order-4 integration method and the JPL97 chemistry. It includes treatment of heterogeneous processes on PSC's and aerosols. The ray tracing model is similar as in SLACO.
- *SLIMCAT1D (University of Leeds):* SLIMCAT1D is the 1D column version of the three-dimensional chemical transport model (3D CTM) SLIMCAT [8]. SCD's are calculated using a single scattering radiative transfer model similar as in SLACO and PSCBOX.

2. Comparison of the photochemical models

In this comparison test, the photochemical models keep their own chemical and photochemical reactions scheme and the heterogeneous processes are switched on. Models are initialised with common SLIMCAT 3D CTM output for the following locations and days: Bremen (53°N, 9°E) 4/3/97 12h UT and Ny-Ålesund (79°N, 12°E) 19/3/97 12h UT.
Examples of photochemical models output for Bremen and for the level corresponding to 20 km of altitude are shown in figures 1-6. Similar figures are obtained for Ny-Ålesund.



Figures 1-6: Examples of photochemical models output for Bremen (53°N) at 20 km of altitude. Legend : X-axis: run time (h); Y-axis: volume mixing ratio ; ——— : PSCBOX ; : BRAPHO ; ------ : SLIMCAT1D.

Concerning BrOy species, a good agreement between the models is observed for BrO whereas discrepancies are observed for $BrONO_2$, BrCl (not shown) and HOBr. The discrepancies observed during night for the last three species are due to differences in the treatment of bromine heterogeneous chemistry mainly between PSCBOX on one hand and SLIMCAT1D and BRAPHO on the other hand. In the case of HOBr, the discrepancies observed during daylight between BRAPHO and the two other models can not be due to the HOBr cross sections used since they are the same in SLIMCAT1D and BRAPHO (Ingham et al. [9] ; JPL97 in PSCBOX). So, more investigations, e.g. comparison of the reactions scheme, are needed to explain this result.

In the case of BrO, a good agreement between the three models is observed for levels between 12 km and 45 km of altitude. More generally, for most of the chemical species the agreement between the photochemical models depends on the altitude: a good agreement is observed at some altitudes but discrepancies appear at others.

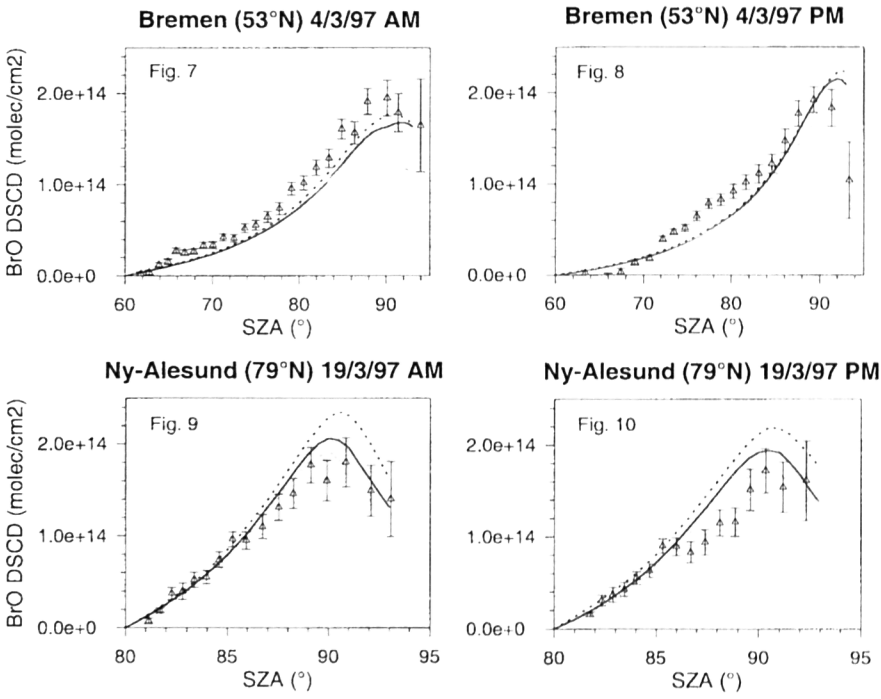
These discrepancies could be due to differences in the reactions schemes, in the photodissociation coefficients and in the heterogeneous chemistry processes.
Comparison tests with common reactions scheme, common photochemical and kinetics data and heterogeneous processes switched on and off are currently under progress in order to find the origins of the discrepancies between the models output.

3. Comparison of simulated BrO SCD's:

Each group initialises its ray tracing model with its own photochemical model output and BrO SCD's are calculated for both locations. The conditions of initialisation of the ray tracing models are not imposed in this comparison test. They are the following:

- SLACO: 0-60 km/1 km as altitude grid, BrO set to zero below 12 km of altitude, p,T profiles from UKMO, O3 absorption and aerosols scattering and refraction included, wavelength: 350 nm
- PSCBOX: 0-70 km/0.2 km as altitude grid, BrO is set to zero below 12 km and above 50 km of altitude, p,T profiles from MPI, O3 absorption included, aerosols scattering and refraction not included, wavelength: 352 nm
- SLIMCAT1D: 0-70 km/1 km as altitude grid, BrO is set to zero below 12 km and above 60 km of altitude, p,T profiles from MPI, O3 absorption and aerosols scattering and refraction not included, wavelength: 352 nm

Simulated BrO differential SCD's (DSCD's) are compared to measurements in figures 7-10.



Figures 7-10: BrO DSCD's (SCD's at SZA - SCD's at minimum SZA of the day) as a function of the solar zenith angle (SZA). Legend : Δ : measurements ; ——— : PSCBOX ; : BRAPHO ; ------: SLIMCAT1D.

For both locations SLIMCAT1D values are larger than PSCBOX and SLACO values especially at large solar zenith angles. Possible reasons are: the use of different altitude grids and number of levels initialised with BrO, O3 absorption which is not included in the SLIMCAT1D ray tracing model and larger BrO amounts at high altitudes in SLIMCAT1D photochemical model output.
Since discrepancies are observed between simulated BrO DSCD's, it is useful to compare into more details the ray tracing models (e.g. comparison of the interpolation routines) and to determine the impact of the differences in the conditions of initialisation on the calculated BrO SCD's. These tasks are currently under progress.
Considering the quite large uncertainties of the measurements at both locations, the three models reproduce reasonably well the measurements for Ny-Ålesund whereas discrepancies are more severe for Bremen especially at large solar zenith angles. Comparisons for more than two days are needed to conclude on the agreement between measurements and models. A comparison of BrO DSCD's simulated from 3D CTM SLIMCAT with measurements performed at a global network of ground-based sites is presented by Sinnhuber *et al.* [10].

4.Conclusions

At this stage of the intercomparison exercise the following conclusions can be drawn:

- For all the chemical species the agreement between the photochemical models is reasonably good even if discrepancies appear at some altitudes for some species. In order to find the origins of these discrepancies, comparison tests with common reactions scheme, common photochemical and kinetics data and heterogeneous chemistry processes switched on and off are currently under progress.
  Concerning BrO, a good agreement is observed for all the levels up to 45 km of altitude.
- Discrepancies are observed between simulated BrO SCD's. These discrepancies could be due to the differences in the conditions of initialisation of the ray tracing models but also to the models themselves (e.g. possible differences in the interpolation routines). The impact of these differences on the simulated BrO SCD's are currently being determined.
  The three models reproduce reasonably well the measured BrO DSCD's for Ny-Ålesund whereas discrepancies are more severe for Bremen but comparison for more than two days is needed to conclude on the agreement between the measurements and the simulations.

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