Comparison of ground-based BrO measurements during THESEO with the SLIMCAT chemical transport model

B.-M. Sinnhuber,[1] M.P. Chipperfield,[1] C.-F. Enell,[2] U. Frieß,[3] F. Hendrick,[4] P.V. Johnston,[5] K. Kreher,[5] K. Pfeilsticker,[3] U. Platt,[3] A. Richter,[6] A. South,[7] K. K. Tørnkvist,[8] M. Van Roozendael,[4] T. Wagner,[3] F. Wittrock[6]

1. School of the Environment, University of Leeds, United Kingdom
2. Swedish Institute of Space Physics, Kiruna, Sweden
3. Institute of Environmental Physics, Heidelberg University, Germany
4. Belgian Institute for Space Aeronomy, Bruxelles, Belgium
5. National Institute of Water and Atmospheric Research, Lauder, New Zealand
6. Institute of Environmental Physics, University of Bremen, Germany
7. Centre for Atmospheric Science, University of Cambridge, United Kingdom
8. Norwegian Institute for Air Research, Kjeller, Norway

Abstract

Ground-based UV/visible measurements of stratospheric BrO slant columns are compared with simulations from the SLIMCAT 3D chemical transport model. The measurements have been performed at a global network of ground-based sites in the frame of the European THESEO BrO project. The agreement between observations and model is in general very good: The model reproduces many features of the observations including the seasonal and latitudinal variations.

1. Introduction

Bromine compounds are believed to play an important role in the destruction of stratospheric ozone, both at high and mid-latitudes. During daytime about 50% of the total inorganic bromine content is present in the form of the active species BrO and Br. Thus – unlike chlorine – a large fraction of bromine is available in an active form under all conditions, which explains the importance of bromine for the ozone chemistry, although bromine is over a factor of 100 less abundant than chlorine.

Our aim is to test our current understanding of the stratospheric bromine chemistry by comparing BrO from the SLIMCAT 3D chemical transport model with ground-based UV/visible measurements. Previous comparisons [e.g. 1,2] have been limited to short periods and individual locations. Here we look more at the global picture, with emphasis on the latitudinal and seasonal variability. We compare BrO differential slant column densities (DSCD) at twilight from nine observational sites. The sites range from northern high latitudes (Ny-Ålesund, 79° N, Andøya, 69° N, Kiruna, 68° N) over northern mid-latitudes (Harestua, 60° N, Bremen, 53° N, OHP 44° N, Huelva, 37° N) to southern mid- and high latitudes (Lauder, 45° S, Arrival Heights, Antarctica, 78° S). For the comparison we take DSCD for solar zenith angles of 90° – 80°. In case solar zenith angles of 80° are not reached (as in high latitude winter), the lowest solar zenith angle available is used instead.

2. Model description

The model used in this study is the SLIMCAT 3D chemical transport model, described in detail by Chipperfield [3]. The model was initialized in 1991 and integrated until mid

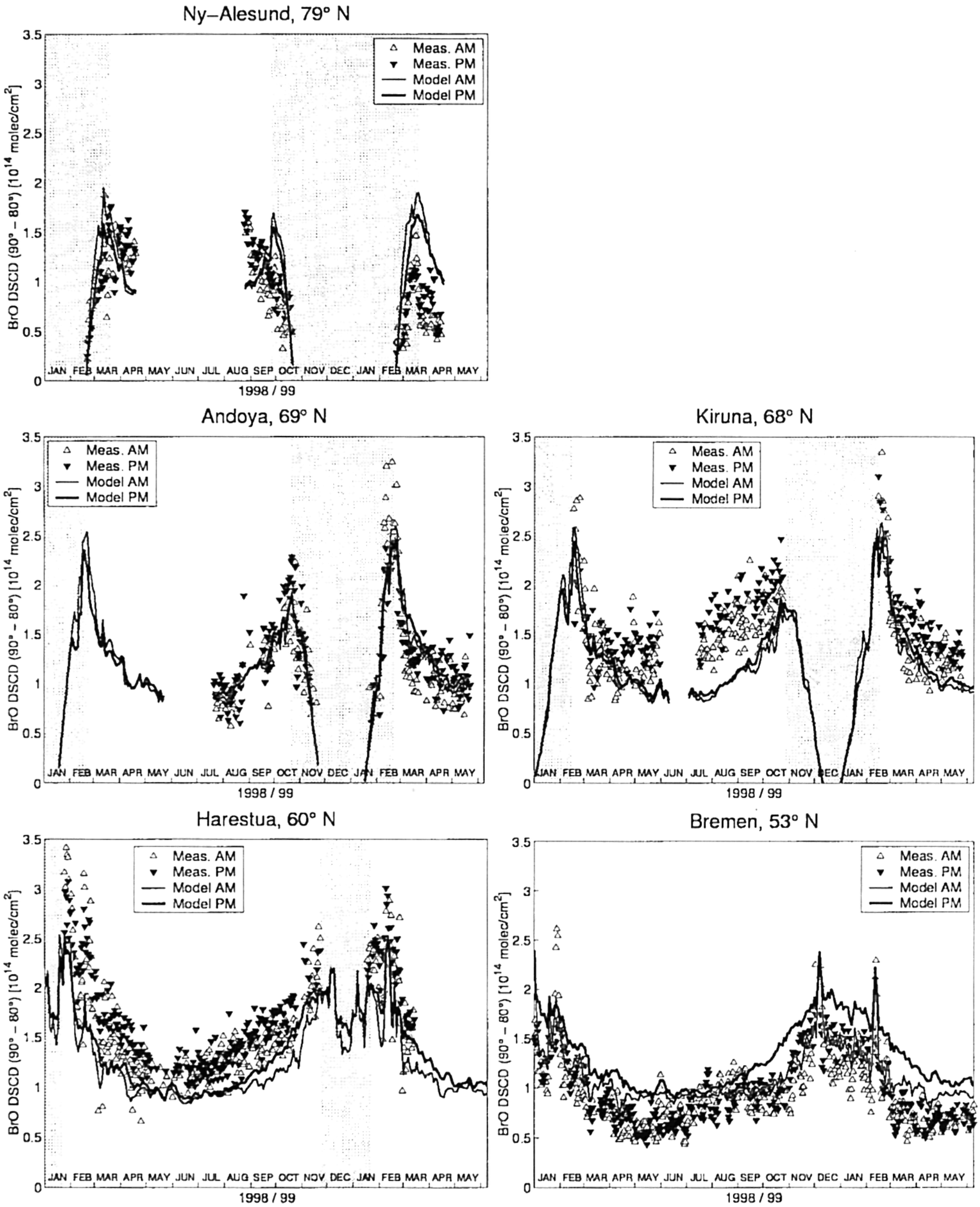

Figure 1. Comparison between observed (triangles) and modeled BrO differential slant column densities (in 10^{14}molecules/cm^2, solid lines). The time series shown range from January 1998 to May 1999. The gray shaded regions indicate periods when the sun did not reach zenith angles of 80°. In this case the lowest solar zenith angle is used for the reference spectrum. Note that some data (e.g. Huelva) are still preliminary.

1999. Photochemical reaction rates and absorption cross sections were taken from JPL97 [4], except for the HOBr absorptions cross section, which was taken from Ingham *et al.* [5].

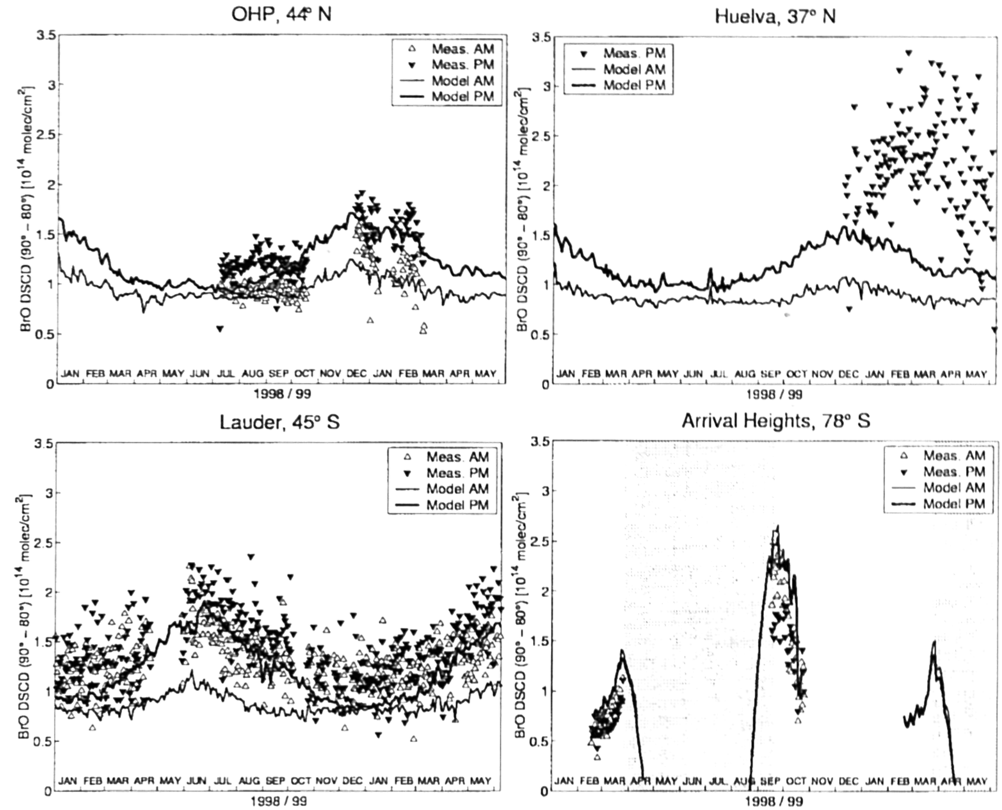

Figure 1, continued.

The model runs shown here assumed a total inorganic bromine loading of 20 pptv.

Differential slant column densities were simulated by coupling the SLIMCAT model to a single scattering ray-tracing radiative transfer model. An intercomparison of this model with two other models is presented by Hendrick *et al.* [6].

3. Discussion

The comparison of the modeled BrO DSCD with the observations is shown in Fig. 1. Generally a good agreement can be seen. The model captures many features of the observations. The latitudinal and seasonal variability, together with the AM/PM difference is well reproduced. Events of high BrO DSCD associated with the advection of polar vortex air with high levels of activated chlorine, most obvious in the Bremen data, are well captured by the model as well. No clear overall systematic difference between the model and the observations can be seen. However, relatively large discrepancies can be seen for individual sites, like e.g. for Harestua or for Kiruna during summer, where the model underestimates the observations. Part of the discrepancies may be attributed to uncertainties in the measurements at the individual sites.

The good overall agreement gives us confidence to use the model for an assessment of the role of bromine in mid-latitude ozone loss. However, it should be noted, that recent laboratory studies [7,8] indicate, that the NOx to NOy ratio can in certain situations be about 30% higher in the lower stratosphere, compared to the model runs presented here. This will then result also in higher $BrONO_2$ concentrations and thus lower BrO concentrations. However, first model calculations show, that BrO will be reduced by only about 5% during daytime, because the increase in $BrONO_2$ is partly compensated by a reduction in HOBr. We will therefore re-run the model with an updated NOy chemistry in the near future.

4. Conclusion

We compared ground-based UV/visible observations of twilight BrO differential slant column densities with the SLIMCAT 3D chemical transport model. The model generally shows a very good agreement with the observations, reproducing many observed features. Given the measurements uncertainties, no clear systematic difference between the model and the observations can be seen. In fact the model can be used as a transfer to compare observations at different locations, to help to identify problems in the observations. The generally good agreement between model and observations gives us confidence to use the model for an assessment of the impact of bromine on mid-latitude ozone loss.

Acknowledgements

This work was funded by the Commission of the European Union and the UK Natural Environment Research Council.

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