The Vertical Distribution of Atmospheric BrO from Ground-Based Measurements

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Abstract

Ground-based UV-Visible measurements targeting BrO were made at Lauder, New Zealand (45.0°S, 169.7°E) and Arrival Heights, Antarctica (77.8°S, 166.7°E). Differential Optical Absorption Spectroscopy (DOAS) was used to determine differential slant column densities (DSCDs) from the radiance measurements. UV-Visible measurements have been made in the two complementary viewing geometries of direct-sun and zenith-sky.

A spherical curved earth single scattering radiative transfer model was developed. The effects of refraction, molecular absorption, Rayleigh and Mie scattering were included. Singularity at the tangent point was avoided and a complete intensity calculation performed. The DSCDs for both the direct-sun and zenith-sky viewing measurements were calculated with this forward model.

A general optimal estimation retrieval algorithm was developed to retrieve altitude information by combining DSCDs from the direct-sun and zenith-sky viewing geometries. A complete retrieval characterisation and error analysis was performed. The characterisation illustrated that tropospheric sensitivity was obtained from the direct-sun viewing measurements, while the zenith-sky measurements were essential for stratospheric sensitivity.

Stratospheric and tropospheric BrO columns were retrieved for the diurnal stages of 80°, 84° and 87° SZA for Lauder. The diurnal and seasonal variation of the stratospheric column was successfully retrieved from the measurements. The stratospheric columns were consistent with a stratospheric Br_y loading of 20 ppt. The tropospheric BrO column retrieved over Lauder was less than 0.9 ppt if a uniform distribution throughout the troposphere is assumed. This is consistent with, though lower than, previous estimates of BrO in the free troposphere of 0.5-2.0 ppt (*Richter et al.*, 2002).

The results of a ten week measurement campaign at Arrival Heights for the spring 2002 are presented. Stratospheric and tropospheric BrO columns were retrieved at 80°, 84° and 88° SZA. A high variability was observed for the retrieved stratospheric columns, due in part to the unusual stratospheric warming in the Antarctic spring 2002 (*Allen et al.*, 2003). A mean ubiquitous tropospheric background of 0.3 ppt was retrieved. Also a 'bromine explosion' event was observed, corresponding to a BrO mixing ratio of 7 ppt for a uniformly mixed boundary layer.

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