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Seasonal variation of stratospheric and tropospheric BrO columns derived from ground-based DOAS measurements at 60˚N and 22˚S

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UV/Visible ground-based zenith-sky observations have been performed at Harestua (60˚N, Southern Norway) since 1998 and at Reunion Island (22˚S, Indian Ocean) since August 2004. From analysis of the measured spectra using the Differential Optical Absorption Spectroscopy (DOAS) technique, the slant column of important atmospheric trace gases like O_3, NO_2, BrO, OClO, HCHO, SO_2 can be retrieved. In this study, we focus on the evaluation of bromine monoxide (BrO) data sets. Although tropospheric BrO signatures have been identified at a few polar stations, ground-based DOAS measurements of BrO have been mainly used to infer information on the stratospheric BrO content, based on analysis of twilight differential slant columns. In the present work, the full diurnal variation of the BrO slant columns is exploited to derive independent information on the stratospheric and tropospheric parts of the total BrO column. Measured slant columns are fitted using a validated forward model accounting for (1) the transfer of the multiply scattered radiation in a pseudo spherical atmosphere and (2) the diurnal variation of the stratospheric BrO modeled using recent reaction coefficients (JPL 2000). After inversion of Harestua data, we find stratospheric columns characterized by a symmetric U-shaped seasonal variation with a summer minimum and a winter maximum, roughly consistent with 3D model calculations. Significant and highly variable tropospheric columns (up to 3-4 x10^{13} molec/cm^2) are found in spring, suggesting a contribution due to transport from polar sources. The tropospheric content is minimum in summer (below 1 x10^{13} molec/cm^2). At the tropical site of Reunion Island, the instrument is equipped with a multi-axis system, which increases its sensitivity to the troposphere. From the combined analysis of zenith-sky and off-axis measurements, a mean wintertime tropospheric content of approximately 1x 10^{13}

molec/cm^2 is found, which represents one third of the total column.