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Retrieval of stratospheric and tropospheric BrO profiles and columns using ground-based zenith-sky DOAS observations at Harestua, 60° N

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Abstract. A profiling algorithm based on the optimal estimation method is applied to ground-based zenith-sky UV-visible measurements from Harestua, Southern Norway (60° N, 11° E) in order to retrieve BrO vertical profiles. The sensitivity of the zenith-sky observations to the tropospheric BrO detection is increased by using for the spectral analysis a fixed reference spectrum corresponding to clear-sky noon summer conditions. The information content and retrieval errors are characterized and it is shown that the retrieved stratospheric profiles and total columns are consistent with correlative balloon and satellite observations, respectively. Tropospheric BrO columns are derived from profiles retrieved at 80° solar zenith angle during sunrise and sunset for the 2000–2006 period. They show a marked seasonality with mean column value ranging from $1.52\pm0.62\times10^{13}$ molec/cm² in late winter/early spring to $0.92\pm0.38\times10^{13}$ molec/cm² in summer, which corresponds to 1.0±0.4 and 0.6±0.2 pptv, respectively, if we assume that BrO is uniformly mixed in the troposphere. These column values are also consistent with previous estimates made from balloon, satellite, and other ground-based observations. Daytime (10:30 LT) tropospheric BrO columns are compared to the *p*-TOMCAT 3-D tropospheric chemical transport model (CTM) for the 2002–2003 period. *p*-TOMCAT shows a good agreement with the retrieved columns except in late winter/early spring where an underestimation by the model is obtained. This finding could be explained by the non-inclusion of sea-ice bromine sources in the current version of *p*-TOMCAT. Therefore the model cannot reproduce the possible transport of air-masses with enhanced BrO concentration due to bromine explosion events from the polar region to Harestua. The daytime stratospheric BrO columns are compared to the SLIMCAT stratospheric 3-D-CTM. The model run used in this study, which assumes 21.2 pptv for the Br_y loading (15 pptv for long-lived bromine species and additional 6 pptv for very short-lived species (VSLS) added by a scaling of CH_3Br), significantly underestimates the retrieved BrO columns. A sensitivity study shows that a good agreement can only be obtained if 6

to 8 pptv accounting for VSLS are added directly (and not by a scaling of CH_3Br) to the SLIMCAT long-lived bromine species profile. This contribution of the VSLS to the total bromine loading is also consistent with recently published studies.

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