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Retrieval of stratospheric and tropospheric BrO columns from multi-axis DOAS measurements at Reunion Island (21° S, 56° E)

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Abstract. Spectral measurements of BrO using zenith-sky and off-axis viewing geometries are combined in a linear multiple regression retrieval algorithm to provide stratospheric and tropospheric BrO vertical columns. One year of measurement data are investigated over Reunion-Island (20.9° S, 55.5° E), from August 2004 to June 2005. A comparison between the stratospheric columns retrieved at 45°, 80°, 85°, 87.5° and 92.5° solar zenith angles and photochemical simulations initialized by chemical fields from the 3-D-CTM SLIMCAT and further constrained by observed NO₂ profiles shows a good agreement only by considering a contribution from the very short-lived organic bromine substances to the stratospheric inorganic bromine budget, of 6 to 8 pptv. Furthermore, stratospheric BrO profiles retrieved from late twilight zenith-sky observations are consistent with a total inorganic bromine (Br_y) loading of approximately 23 pptv. This represents 6 to 7 pptv more than can be supplied by long-lived organic bromine sources, and therefore supports an added contribution from very short-lived organic bromine substances as recently suggested in several other studies. Moreover strong evidences are presented for the existence of a substantial amount of BrO in the tropical free-troposphere, around 6 km altitude, possibly supplied by the decomposition of short-lived biogenic bromine organic compounds. Tropospheric BrO vertical columns of $1.1 \pm 0.45 \times 10^{13}$ molec/cm² are derived for the entire observation period. Comparisons between ground-based BrO vertical columns and total BrO columns derived from SCIAMACHY (onboard the ENVISAT satellite) nadir observations in a latitudinal band centered around 21° S present a good level of consistency, which further strengthens the conclusions of our study.

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