



Increase of total stratospheric NO₂ in the tropics after 2001

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The tropical region is the main entry point of tropospheric chemical species lifted by convection and transported into the stratosphere across the tropopause. It is therefore the most sensitive region to dynamical and chemical changes. Long series of NO₂ columns have become available in the tropics from two SAOZ uv-visible spectrometer stations, in Bauru (Brazil, 22°S, 49°W) since 1995 and in Reunion Island (21°S, 55°E) in the Indian Ocean since 1993 with which the evolution of the total NO₂ has been investigated. Most significant modulation at both stations is the seasonal cycle but of larger amplitude $\sim 3.2 \cdot 10^{15} \text{ mol./cm}^2$ in Bauru than $\sim 2.4 \cdot 10^{15} \text{ mol./cm}^2$ in Reunion because of the contribution tropospheric lightning and biomass burning NO_x in continental Brazil, absent over the oceanic Reunion station. As shown by a multi-linear regression analysis, most important geophysical parameters controlling the interannual variability are the Quasi-Biennial Oscillation (QBO) of respectively $\approx 30\%$ in Bauru and 20% in Reunion, followed by the El Niño-Southern Oscillation (ENSO) of 17% and 11% and the 11 year solar flux cycle of 11% at both stations. After removing the signal from all statistically significant proxies, the long-term evolution shows a total NO₂ increase by about $\sim 0.2 \cdot 10^{15} \text{ mol./cm}^2$ after 2001 at both stations, exceeding by far the trend expected from the known N₂O concentration increase of $2.5\%/decade$. The event is confirmed by the GOME - SCIAMACHY NO₂ total column series, but ignored by the SLIMCAT model forced by ECMWF analyses. The partial NO₂ column between 19-50 km derived HALOE and SAGE II above the stations confirms the increase of NO₂ after 2001 and shows that it has occurred in the lower stratosphere. The coincidence of the NO₂ enhancement with the drop of water vapour ($\sim 0.2 \text{ ppmv}$) reported by HALOE in the lower stratosphere and the cooling of $\sim 1\text{K}$ the tropopause, attributed by Randel et al., (2006) to a reinforcement of the Brewer-Dobson circulation, suggests an increase of the NO_x concentration after 2001 following that of its N₂O source, either because of a faster B-D circulation or a more intense convective vertical transport.