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Supporting Information for

Variability of Stratospheric Reactive Nitrogen and Ozone Related to the QBO

M. Park[1], W. J. Randel[1], D. E. Kinnison[1], A. E. Bourassa[2], D. A. Degenstein[2], C. Z. Roth[2], C. A. McLinden[3], C. E. Sioris[3], N. J. Livesey[4] and M. L. Santee[4]

[1]National Center for Atmospheric Research, Boulder, Colorado, USA
[2]Institute of Space and Atmospheric Studies, University of Saskatchewan, Saskatoon, Canada
[3]Air Quality Research Division, Environment and Climate Change Canada, Toronto, Canada
[4]Jet Propulsion Laboratory, Pasadena, California, USA

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Section S1 : Direct Comparison of OSIRIS NO_2 with WACCM4 Results

OSIRIS measures NO_2 number density as a function of altitude, both in descending (morning) and ascending (afternoon) modes. Here we include direct comparisons of OSIRIS NO_2 with WACCM4 simulations sampled like OSIRIS measurements to complement the comparisons of daily average NO_x mixing ratios (NO_x* from OSIRIS) evaluated above. Note that number density is scaled by atmospheric density compared to mixing ratios that are used throughout the rest of this paper, and hence accentuate variations at lower altitudes. OSIRIS NO_2 data here are obtained from descending mode observations only, and after applying a time adjustment factor, are converted to 06:30 LST measurements. The results from the WACCM4 simulations are sampled at OSIRIS measurements locations at 06:30 LST.

Altitude-time sections of the monthly anomalies of OSIRIS NO_2 (06:30 AM) and WACCM4 NO_2 sampled like OSIRIS are shown in Figure S1. Interannual variations of OSIRIS NO_2 reveal strong correlation with the QBO zonal wind anomalies at altitudes between ~28 and 35 km, and show downward propagation to lower altitudes (~20-27 km). The downward propagation is more prominent in number density compared to mixing ratio at lower altitudes as explained above. The WACCM4 simulations show similar interannual variations, and correlations between OSIRIS and WACCM4 NO_2 anomalies are ~ 0.50-0.83 over 46-6.8 hPa with the highest correlation coefficient at 10 hPa. The WACCM4 NO_2 anomalies show larger amplitudes in the upper stratosphere (~28-35 km) compared to OSIRIS NO_2, and this behavior is consistent with the comparisons for daily averaged NO_x mixing ratios discussed above (Fig. 5).

The annual average profiles of OSIRIS NO_2 (06:30AM) and the corresponding WACCM4 result averaged over 5ºS-5ºN latitudes are shown in Fig. S2a. Both the OSIRIS and WACCM4

NO_2 profiles show minima at low altitudes and maxima near ~32 km. OSIRIS NO_2 number density is smaller than WACCM4 at all altitudes between 16-38 km, with differences (Fig. S2b) ranging from ~ 10 to 45%. The best agreement between OSIRIS and WACCM4 (differences smaller than 25%) is found between 23 and 35 km. We note that these differences for time-averaged NO_2 (near 06:30) are slightly larger than for the corresponding daily average NO_x (Fig. 2b), which may suggest some small differences between the detailed NO_x diurnal variations in WACCM4 results and those estimated by the photochemical box model applied to OSIRIS data.

The applicability of the box model in converting NO_2 to NO_x ultimately boils down to how well it can simulate the NO_y species. This was evaluated, both absolutely and in terms of the NO_x/NO_y ratio, through comparisons with observations from 10 flights of the JPL MkIV FTIR (Fourier Transform InfraRed) interferometer [Toon, 1991] between 1997 and 2005 (see Appendix in Brohede et al., 2008). The tropical and mid-latitudes profiles all compared very well, with the only significant discrepancies occurring for a couple of profiles near the polar night.

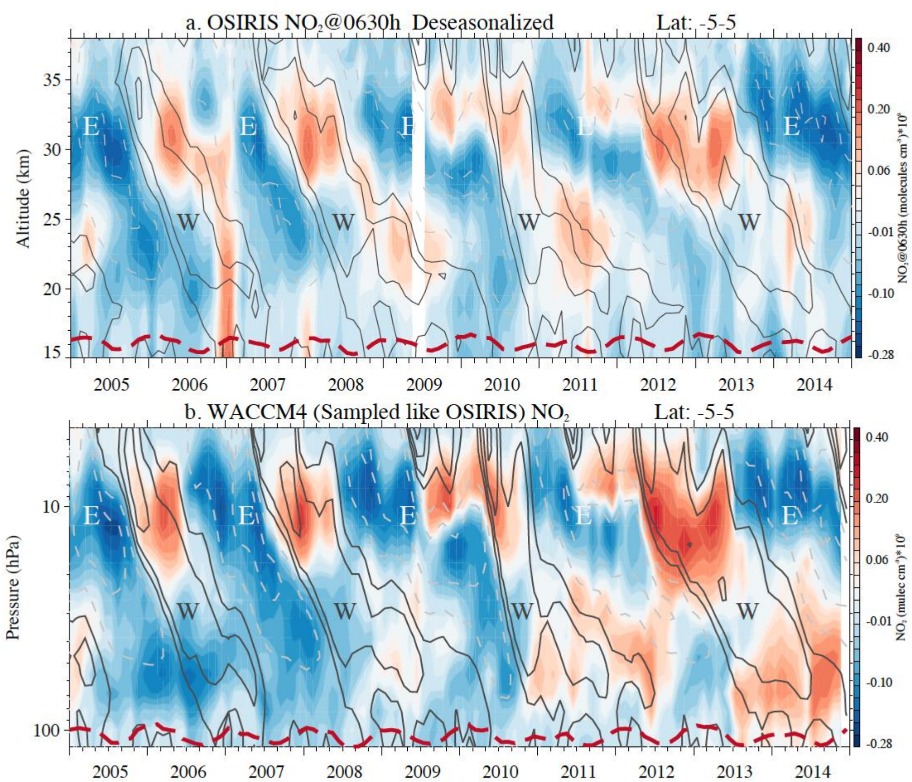


Figure S1: Time versus altitude (pressure) sections of monthly mean anomalies of NO_2 number density at 6:30 AM LST (unit: molecules cm^{-3}) from (a) OSIRIS and (b) WACCM4 sampled like OSIRIS. Data are averaged between 5°S-5°N latitude.

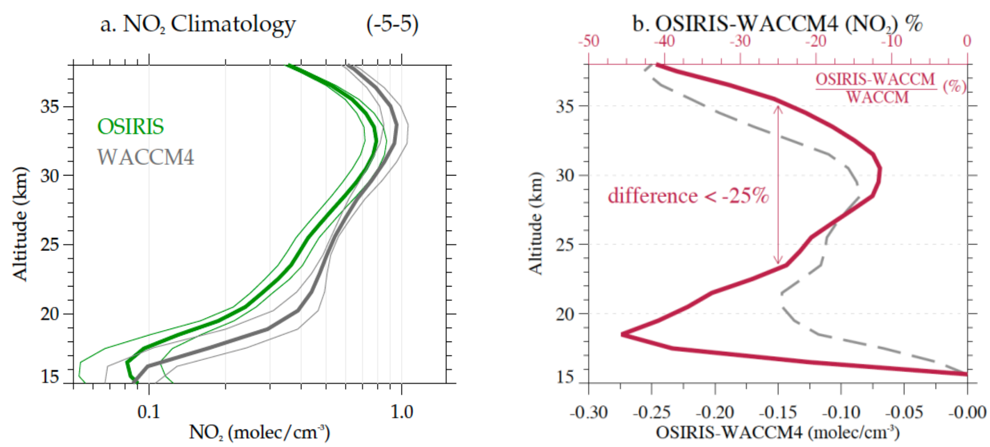
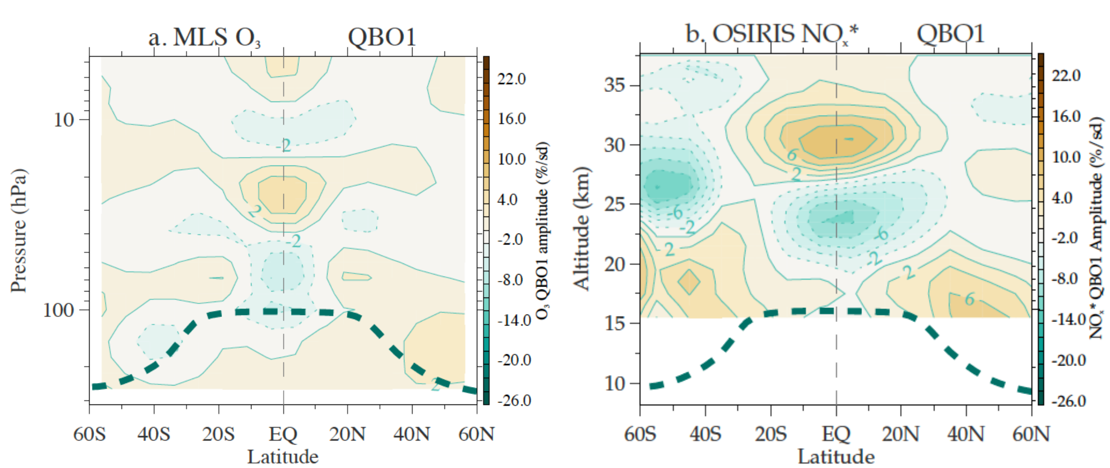


Figure S2. a) Time-averaged vertical profiles of NO_2 number density from OSIRIS (06:30 AM LST, green thick line) averaged between 5°S-5°N latitudes compared with corresponding WACCM results (gray thick line). Thin lines denote one-sigma standard deviation. b) Relative (percent, red solid line) and absolute (gray dashed line) differences between OSIRIS-WACCM4 vertical profiles.

Section S2 : Global QBO regression fits for satellite observations

We have analyzed global QBO regression fits for all of the species studied here, including O_3, N_2O, HNO_3 and NO_x. These fits are based on two orthogonal QBO basis functions (QBO1 and QBO2), as described in the main text, and results are shown for each species in Fig. S3. All species show coherent QBO variations centered in the tropics, along with out-of-phase patterns in the extratropics. QBO1 and QBO2 patterns are spatially orthogonal, representing oscillating QBO variability. Spatial patterns in NO_x and HNO_3 in Fig. S3 are in-phase with each other and out-of-phase with N_2O, and patterns in O_3 and NO_x are also out-of-phase with each other in the upper stratosphere, reflecting the QBO variations in chemical species discussed throughout this paper. Similar calculations for WACCM4 reveal very similar spatial patterns and magnitudes in all cases and are not shown.



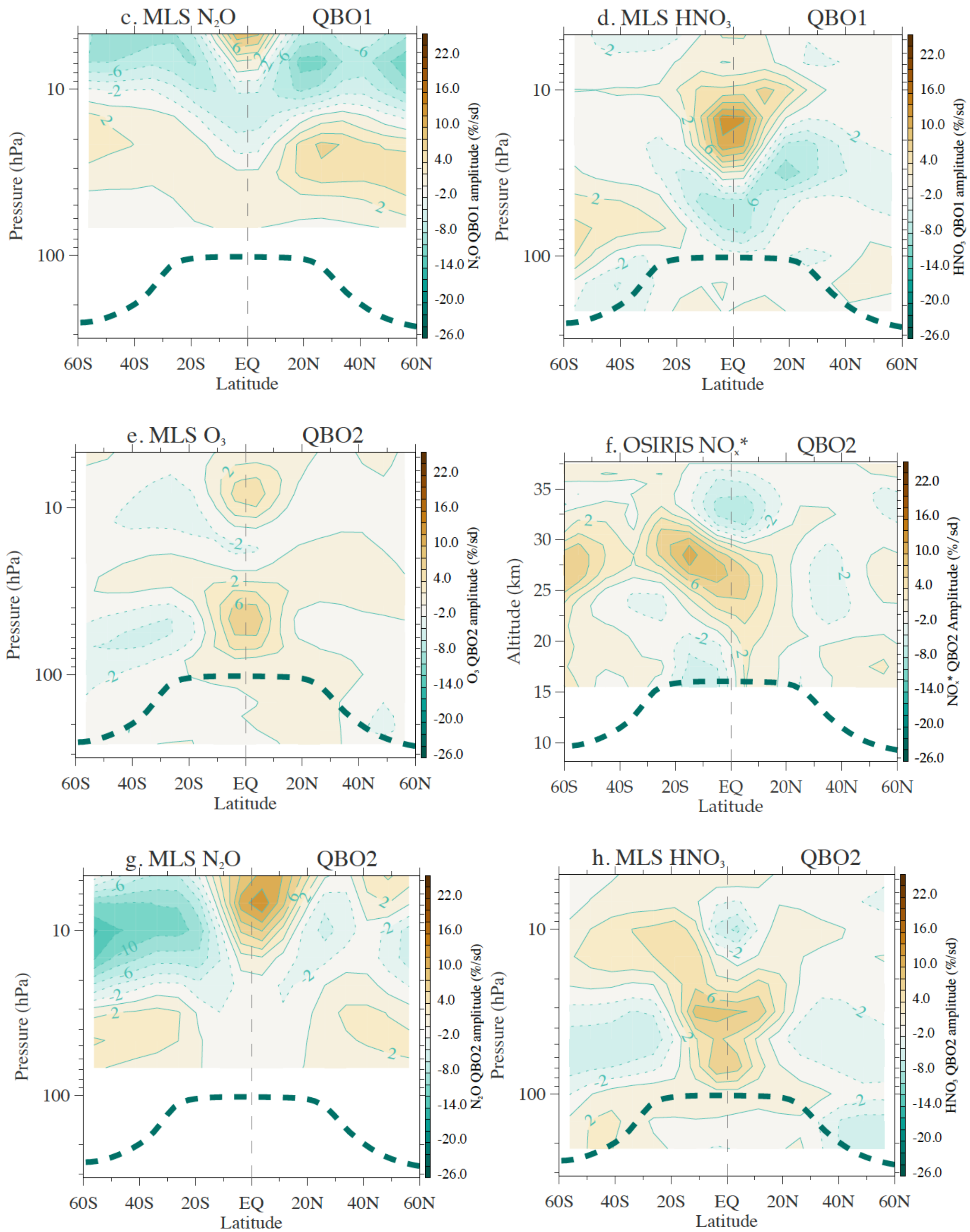


Figure S3. Latitude vs. altitude cross sections of relative amplitude of (a-d) QBO1 and (e-h) QBO2 regression fits for MLS O_3, OSIRIS NO_x*, MLS N_2O and HNO_3. Contours show local percent variations in the respective species associated with one standard deviation of the QBO1 or QBO2 reference time series.