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Supporting Information for

Faster Tropical Upper Stratospheric Upwelling Drives Changes in Ozone Chemistry

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Introduction

This file contains figures that provide information on model-calculated chemical loss processes, observed and simulated trace gas distributions, and correlations between dynamical fields and the observed N_2O. The method for calculating ACE annual mean NO_x data is described.

Details of ACE NO_x Data Usage

We use ACE V4.1 zonal monthly mean data in 5-degree latitude bins on pressure levels. Annual means of ACE NO_x at polar and tropical latitudes were calculated by summing sunrise and sunset measurements of NO, NO_2, and N_2O_5 (x2) in each month available. ACE is an occultation measurement, obtaining roughly 30 profiles daily at a narrow range of latitudes. The 60-70^{o}N/S latitude bands have very good temporal coverage, and each year there are 7-8 months of data. Tropical data from all available months (February, April, August, and October) are used for the annual mean. We calculate the high latitude annual means as the average of the winter average (e.g., calculated using Nov-Mar in the NH) and the summer average (e.g., using May-Sep in the NH). The minimum and maximum of the NO_x annual cycle occurs in these seasons, according to GMI CTM simulations. Based on these simulations, the average of these 2 ACE seasonal means fairly represents the annual mean. Annual means are calculated for consecutive 12-month periods beginning June 2005. The annual means are used in the calculation of the quasi-decadal change.

Figures S1-S9

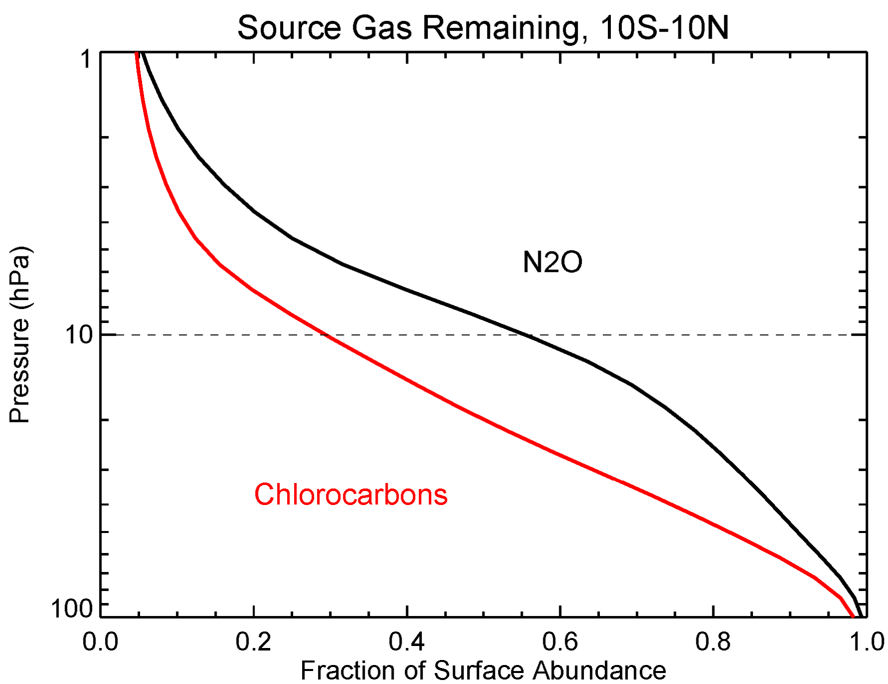


Figure S1. GMI Baseline tropical profiles of source gases normalized to their surface mixing ratios. At 10 hPa, about 70% of chlorine from CFCs and other organic chlorine-containing gases has been released while about 55% of N_2O has not yet been destroyed.

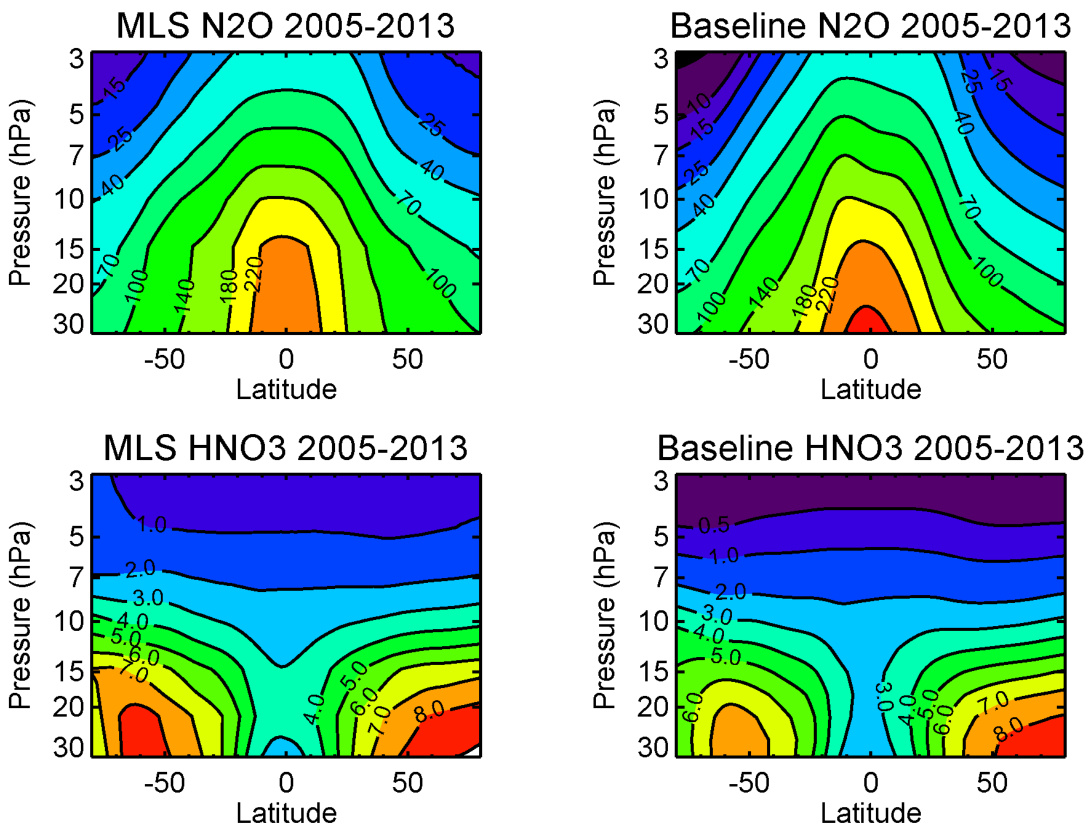


Figure S2. Zonal means averaged 2005-2013 for MLS N_2O and HNO_3 (left column) and GMI Baseline N_2O and HNO_3 (right column), illustrating their latitude-height distributions. For the observations and the simulation, both species have low mixing ratios in the polar upper stratosphere. Errors or uncertainties these low values increase the uncertainty in their quasi-decadal (QD) percentage change above ~7 hPa.

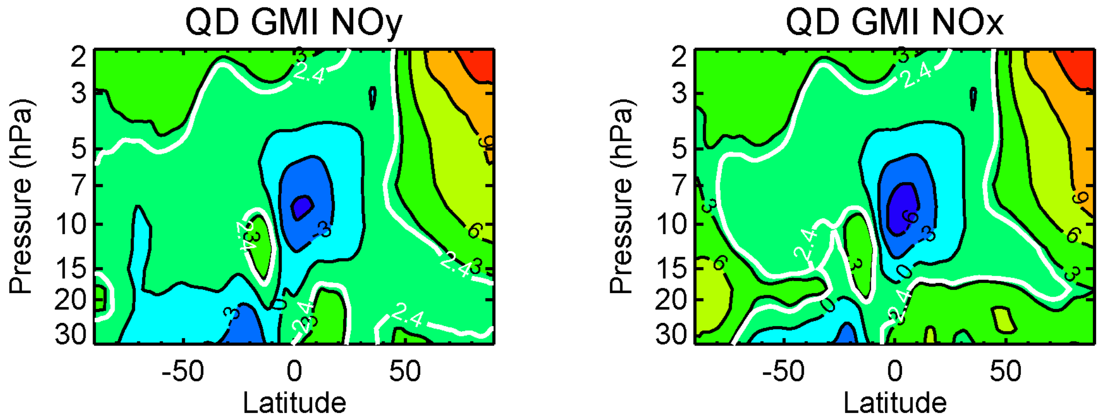
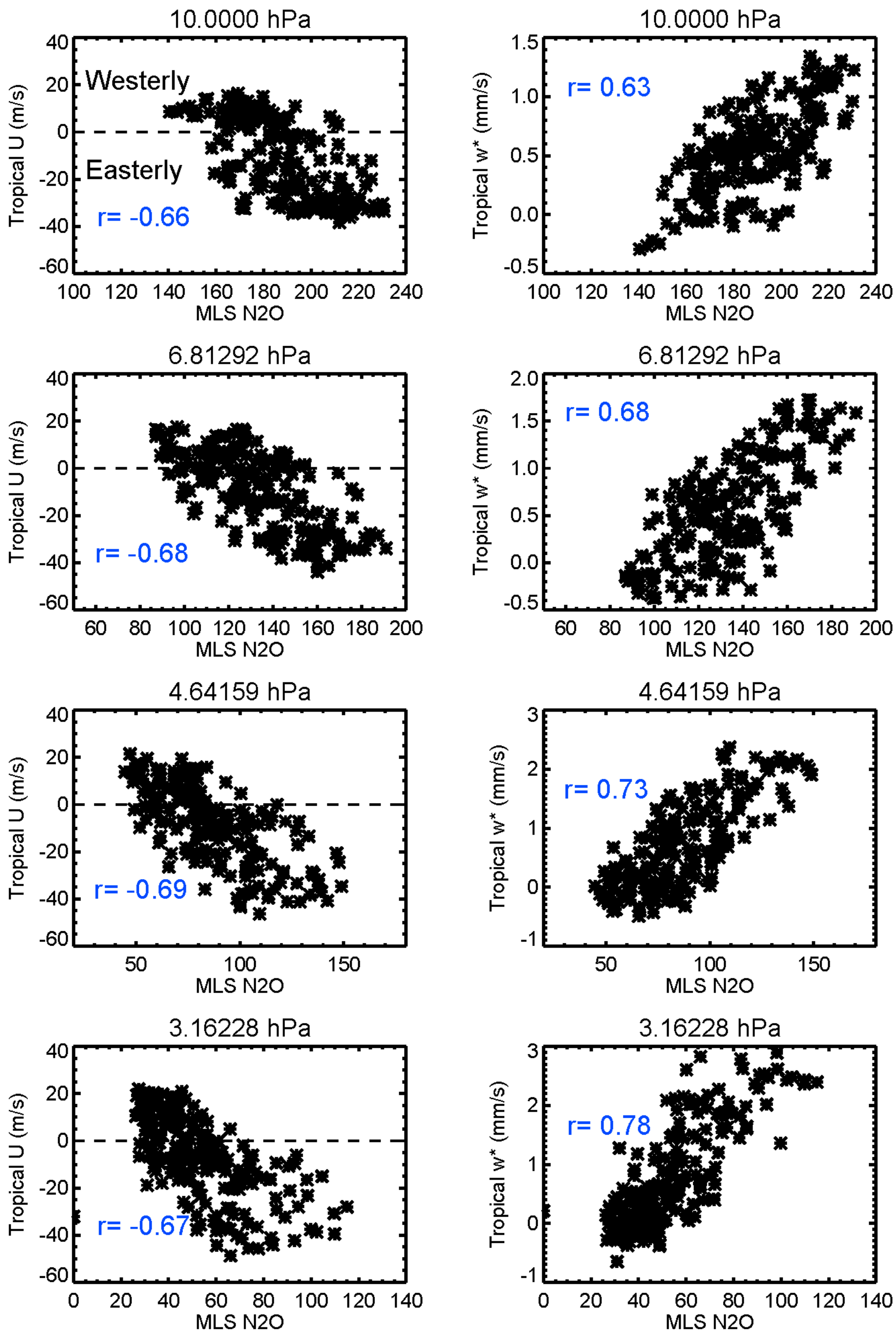


Figure S3. Quasi-decadal (QD) percentage changes in GMI Baseline NO_y (left) and NO_x (right). White contour value, 2.4, is the tropospheric N_2O growth. The patterns of QD changes shown here are similar to those of observed and simulated HNO_3 (Figure 1).



Figures S4. Scatterplots of 10°S-10°N monthly zonal mean MLS N_2O and MERRA2 zonal wind (left column) for June 2005-May 2021. Right column is MLS N_2O and MERRA2 residual vertical velocity, w*. Correlation coefficients are given in each panel.

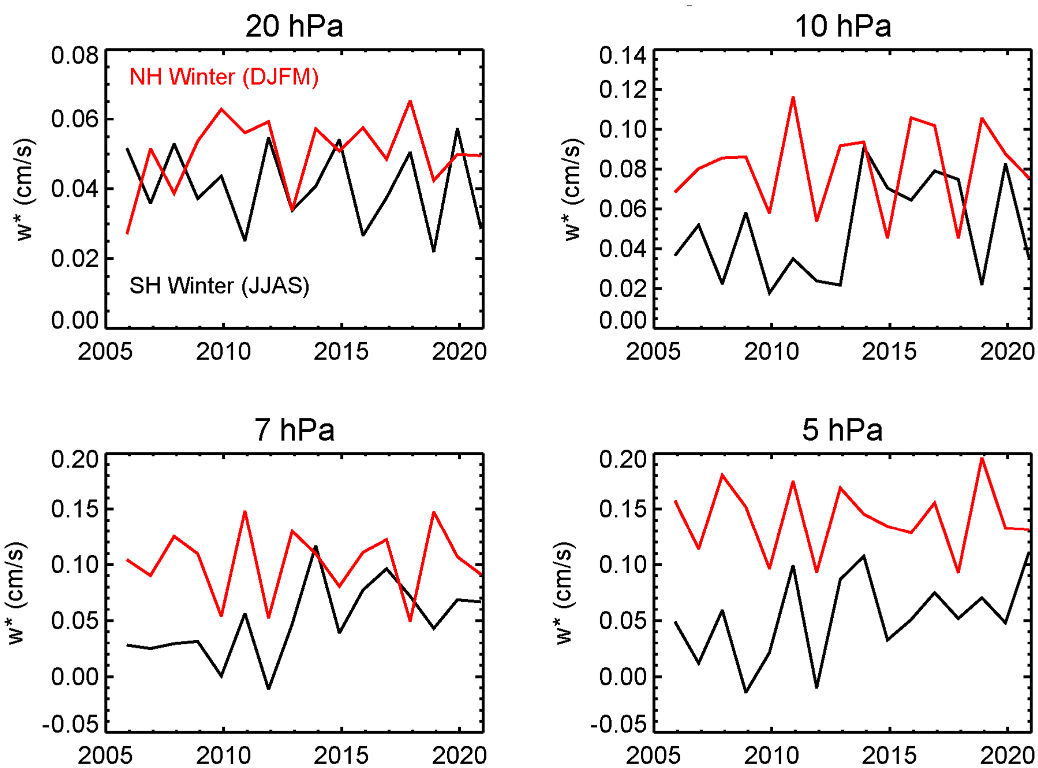


Figure S5. MERRA2 tropical residual vertical velocities (w*) on 4 pressure surfaces, 20-5 hPa, averaged over SH winter months June-September (black) and for NH winter months Dec-Mar (red). w* in NH winters has large interannual variability (IAV) but no obvious trend from 2005-2021. w* in SH winters also has large IAV but is visibly higher on average after 2012 between 10-5 hPa.

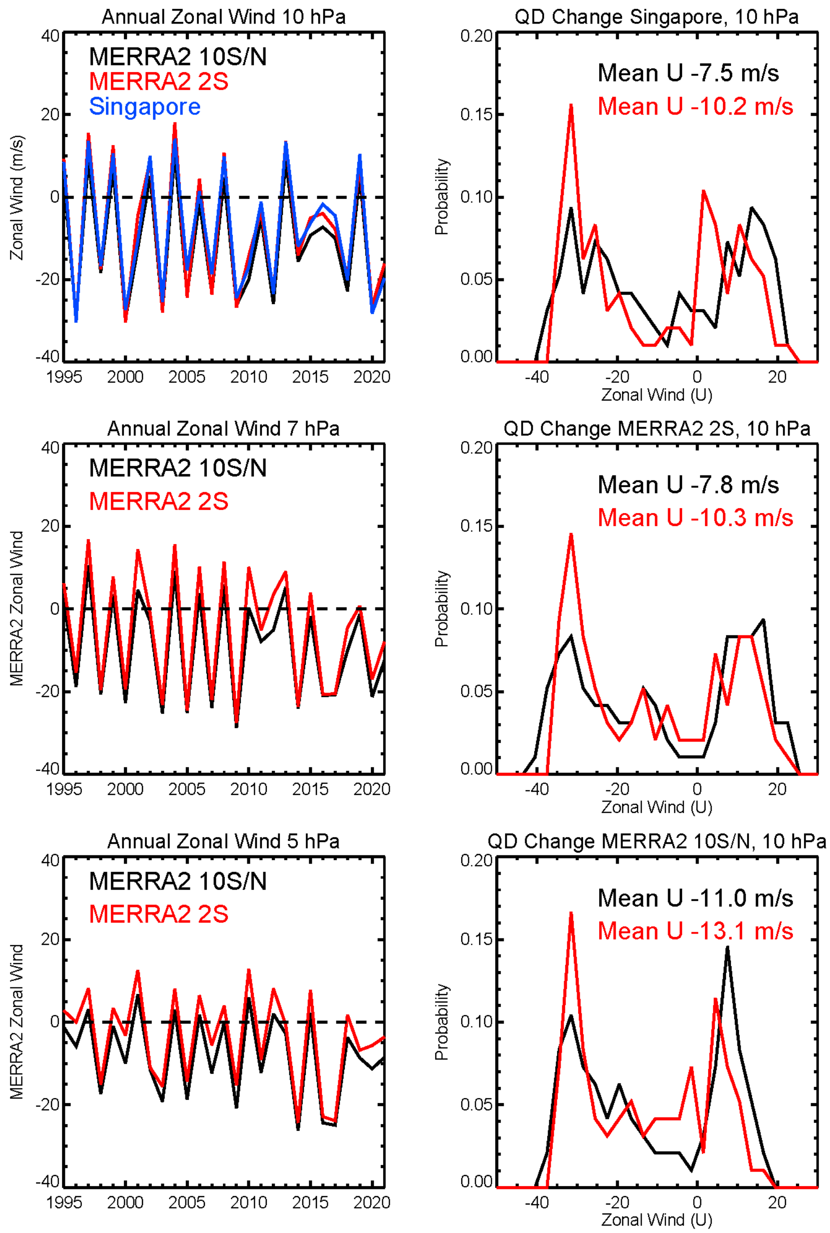


Figure S6. (Top left) Annual means of Singapore (1.3°S) zonal wind data at 10 hPa (blue) and MERRA2 zonal mean zonal wind at 2°S (red) and the 10°S-10°N (tropical) average (black), 1995-2021. The mean difference between Singapore and MERRA2 2°S annual mean winds is less than 1 m/s; the mean difference with the MERRA2 tropical means is -3 m/s, with MERRA2 winds more easterly. (Middle left) MERRA2 zonal winds 1995-2021 at 7 hPa. (Bottom left) MERRA2 zonal winds at 5 hPa. (Right column) 10 hPa distributions of monthly mean Singapore winds (top), MERRA2 2°S winds (middle) and MERRA2 tropical mean winds (bottom) for June 2005-May 2013 (black) and June 2013-May 2021 (red). All show a shift to more easterly winds in the latter period.

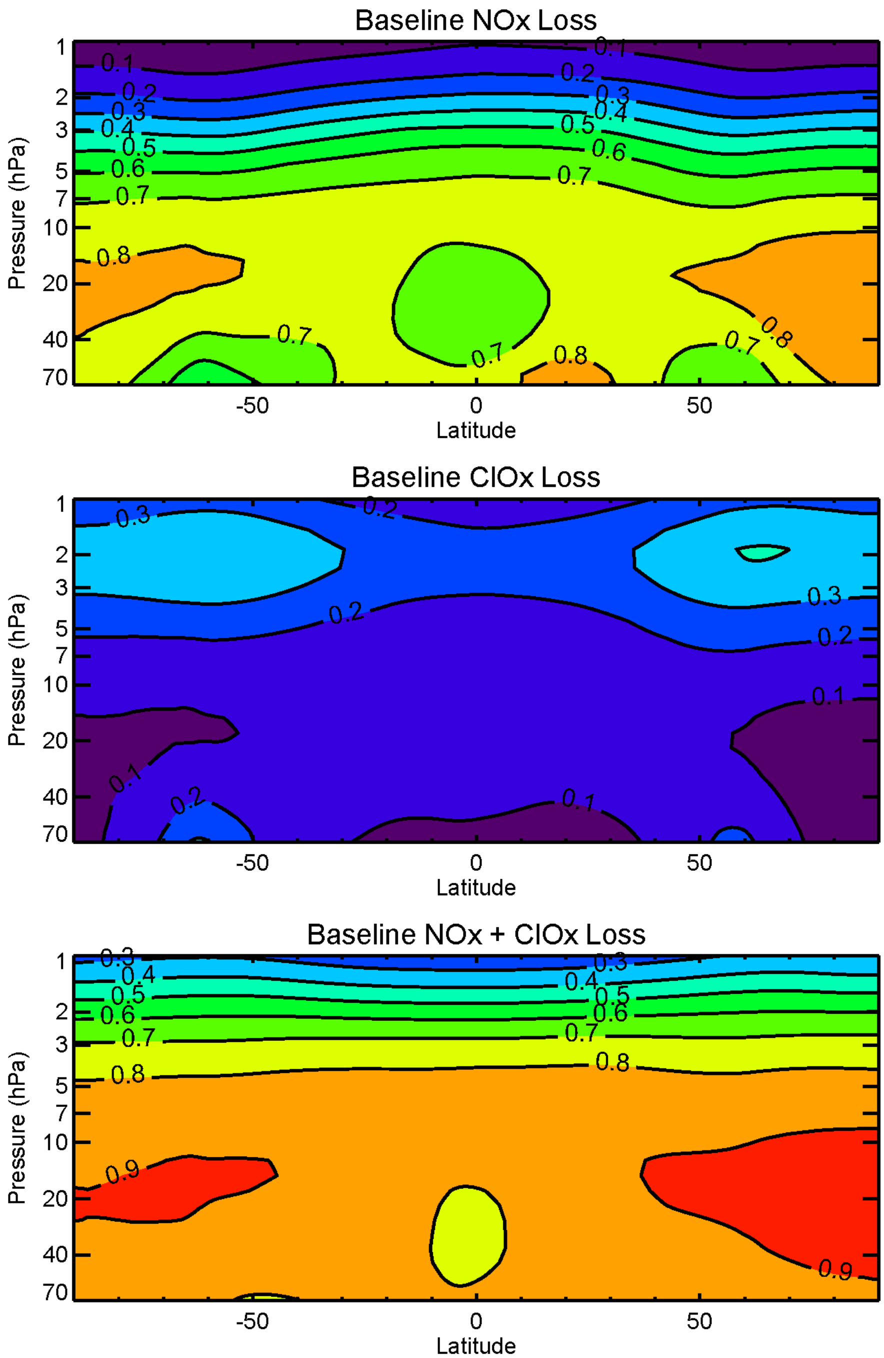


Figure S7. GMI Baseline fraction of total O_3 loss by a) NO_x cycle, b) ClO_x cycle, and c) NO_x+ClO_x. Values are averages over June 2005-May 2013.

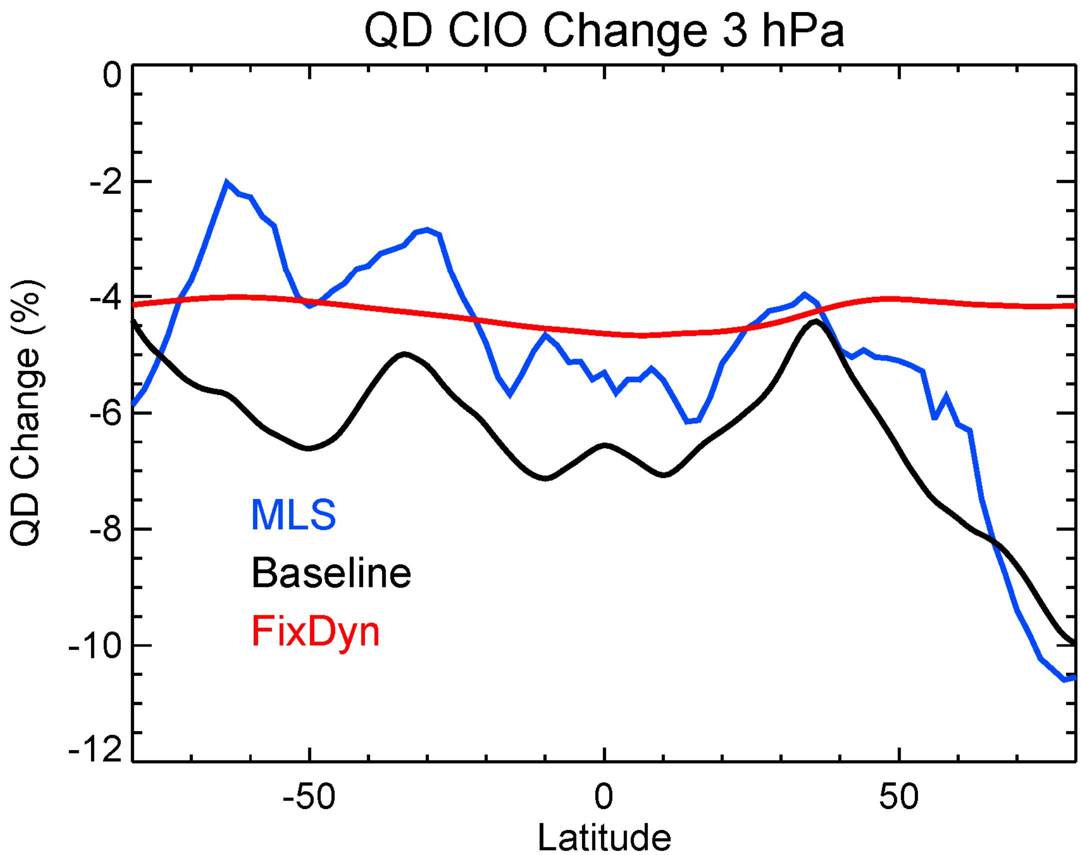
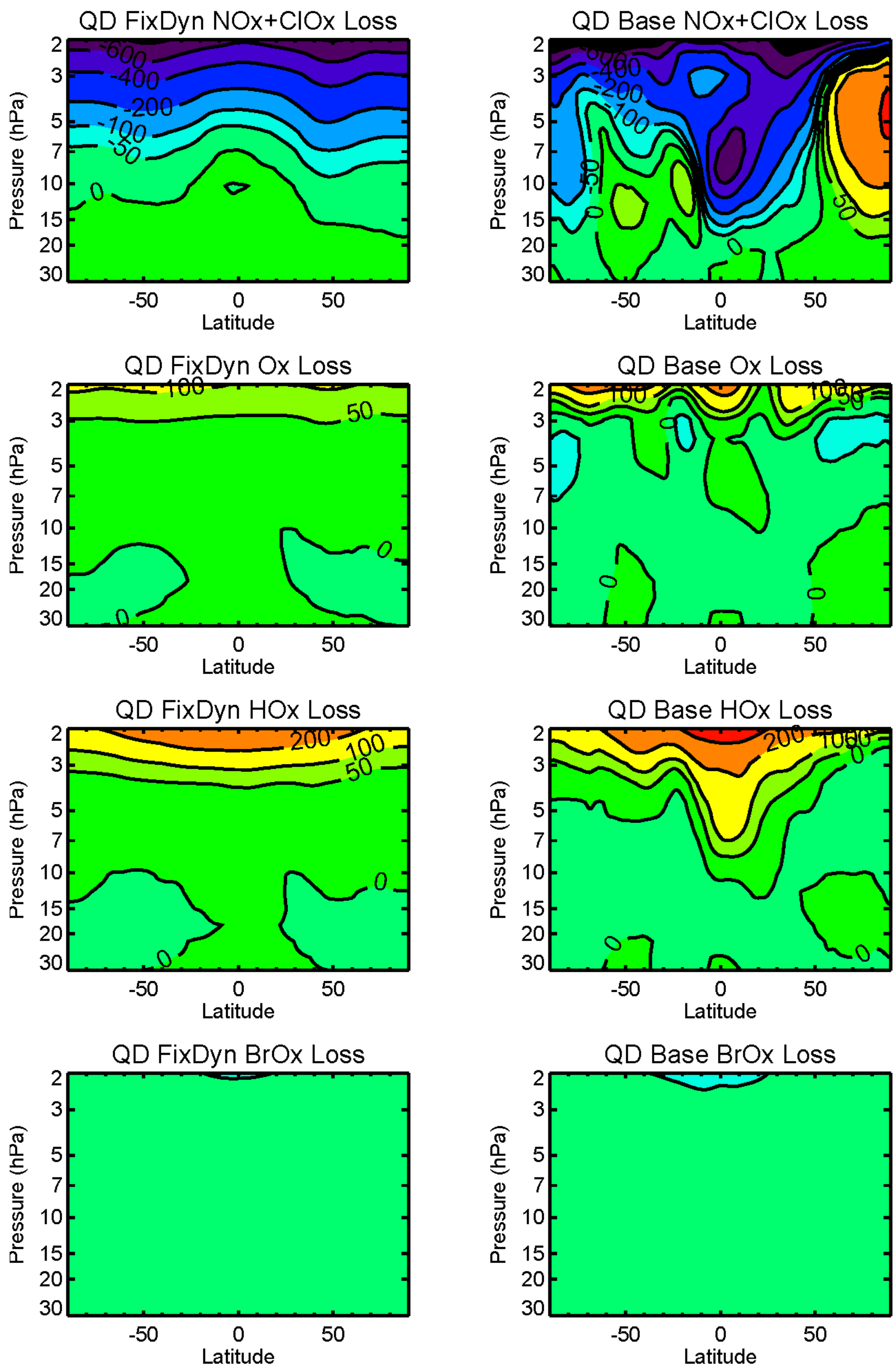


Figure S8. QD changes in ClO for MLS (blue), GMI Baseline (black), and GMI FixDyn (red) at 3 hPa. Baseline ClO decreased more than FixDyn at all latitudes, and both are similar to MLS south of 40°N. North of 50°N, both MLS and Baseline show dramatic decreases in ClO approximately double that of other latitudes. This is caused by the increased Arctic UpS NO_x (see Section 4 of article).



Figures S9. QD changes in all O_3 loss cycles (ppb/mon) for FixDyn (left column) and Baseline (right column). Top row is loss by NO_x+ClO_x, 2nd row is loss by O_x, 3rd row is loss by HO_x, and bottom row is loss by BrO_x. The lowest contour is -800 (deep purple) and the highest is 400 (red).