SUPPLEMENTARY FIGURES

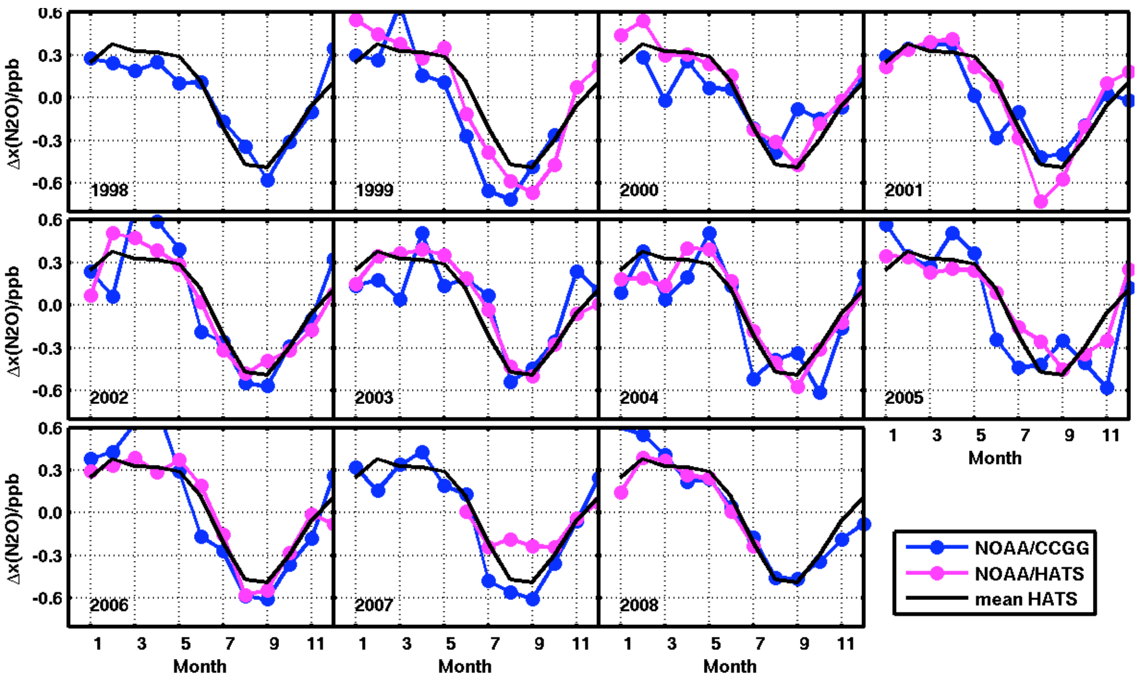

Figure S1) Monthly mean detrended N_2O residuals from NOAA/CCGG and NOAA/CATS networks at Barrow, Alaska.

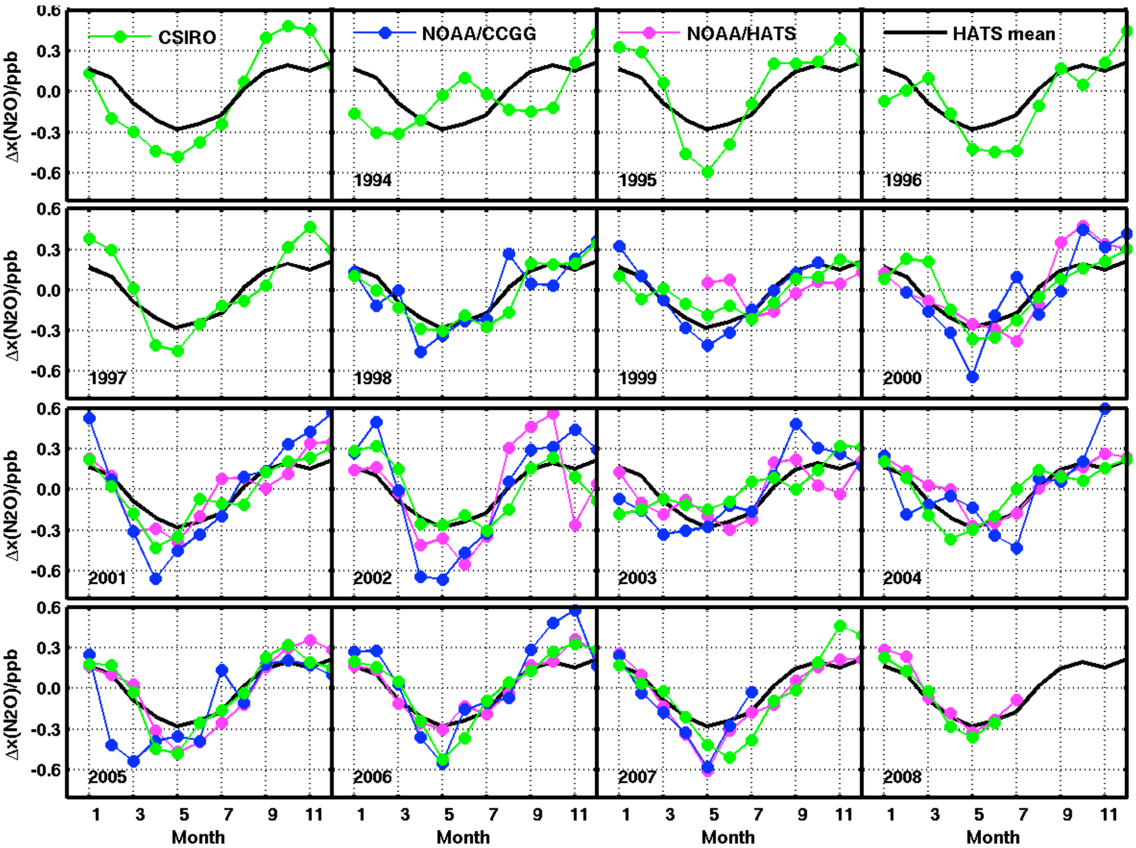

Figure S2) Monthly mean detrended N_2O residuals from CSIRO and NOAA/CCGG and NOAA/CATS networks at the South Pole.

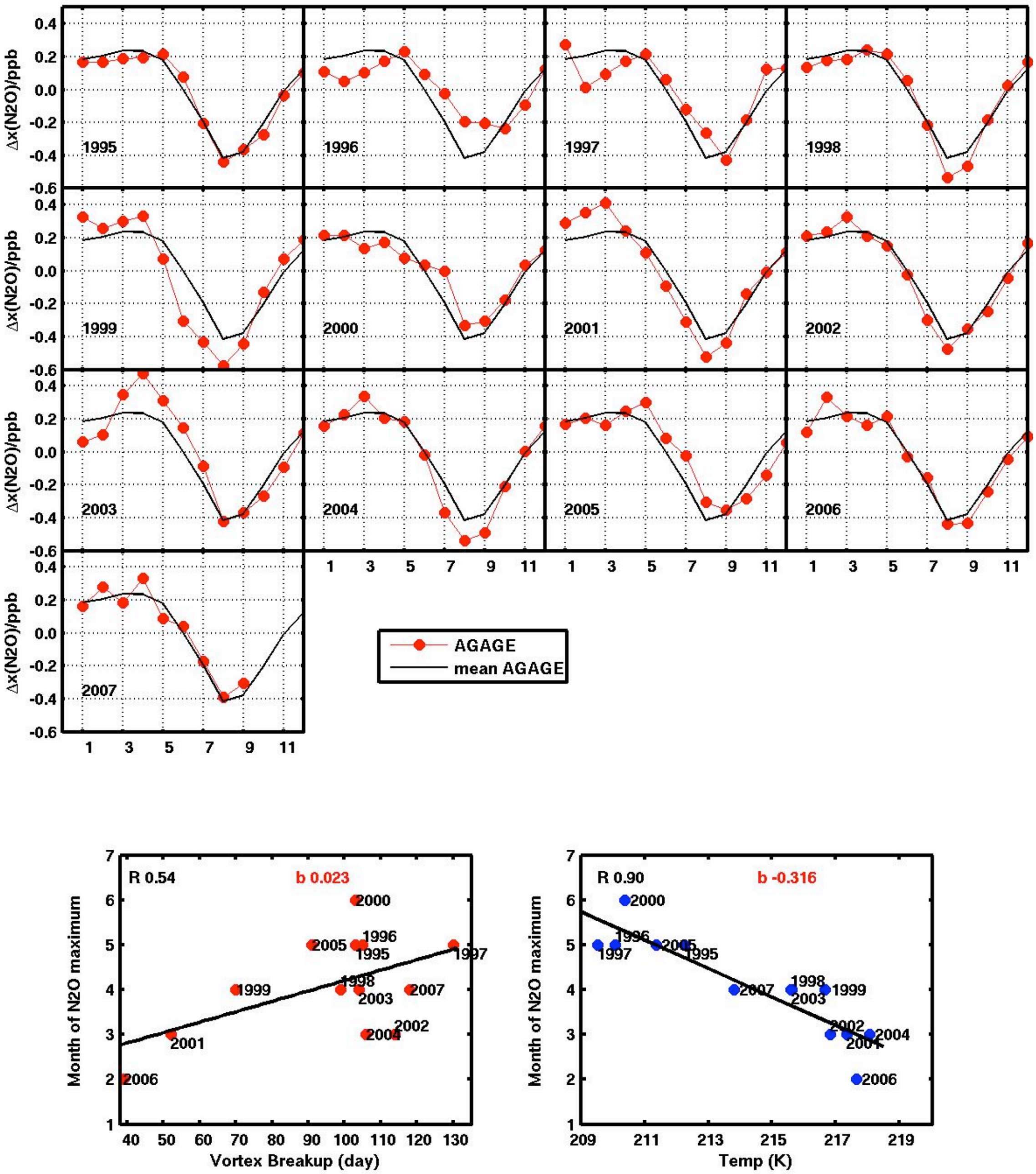

Figure S3. (**Top Panels**) Monthly mean detrended N_2O residuals from AGAGE network at Mace Head, Ireland (repeated from Figure 1a). **(Lower Panels)** Month of onset of descent into summer minimum (referred to as the month of the N_2O maximum) plotted vs. polar vortex breakup date **(Lower Left)** and mean polar (60-90ºN) winter (Jan-March) lower stratospheric (100 hPa) temperature **(Lower Right)**. The Pearson's correlation coefficient *R* and the slope *b* of each linear regression are shown. The N_2O seasonal cycle at MHD is characterized by mainly by its minimum; the month of the maximum is a less distinct feature of the data. We determine the “maximum” by visual inspection based on the onset of a sustained and steep descent into the summer minimum. This definition is clear in most years from 1995-2007, but is somewhat subjective in 1998, 2000, and 2006. When alternate choices are used in those years, R drops from 0.54 to 0.10 (vs. vortex breakup) and from 0.9 to 0.71 (vs. stratospheric temperature). Similar regressions were performed for N_2O and CFC-12 at the other northern sites in Table 1. Quantitatively similar correlations between the NOAA/CCGG N_2O maximum at MHD and stratospheric temperature were found (b= -0.310, R=0.57). For all other datasets, (including AGAGE CFC-12 at MHD), no correlations were found, in part due to ambiguity and relatively large uncertainty in the identification of the month of the seasonal maximum (i.e., onset of descent into the minimum).

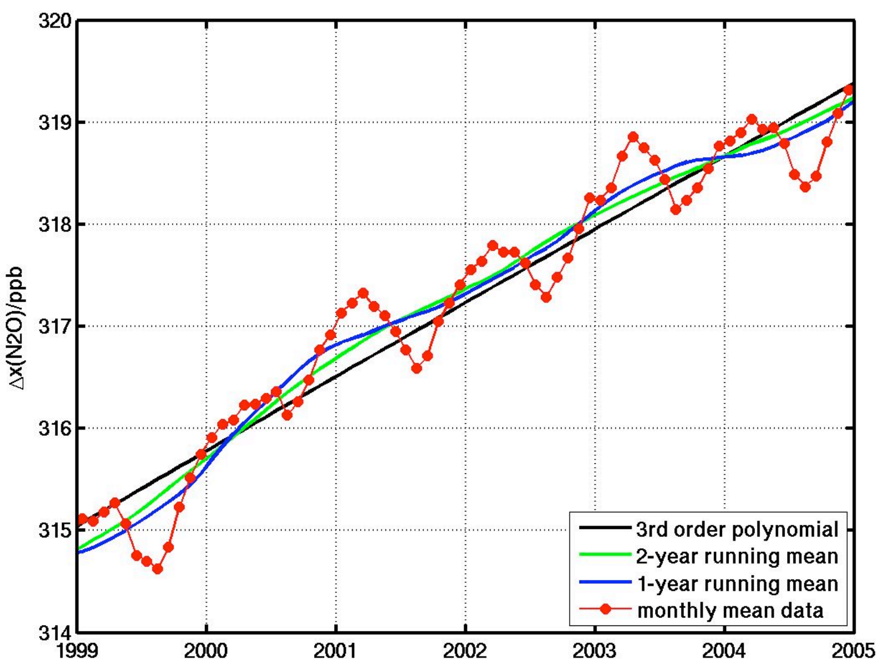

Figure S4. AGAGE monthly mean N_2O data at Mace Head, showing three possible detrending curves. The standard curve used in this paper is the 1-year running mean (blue), which best removes the low frequency variability from the data, allowing interannual changes in the seasonal cycle to be isolated. The detrended monthly means used in the regressions against polar lower stratospheric temperature represent the difference between the monthly mean data (red circles) and the detrending curve.

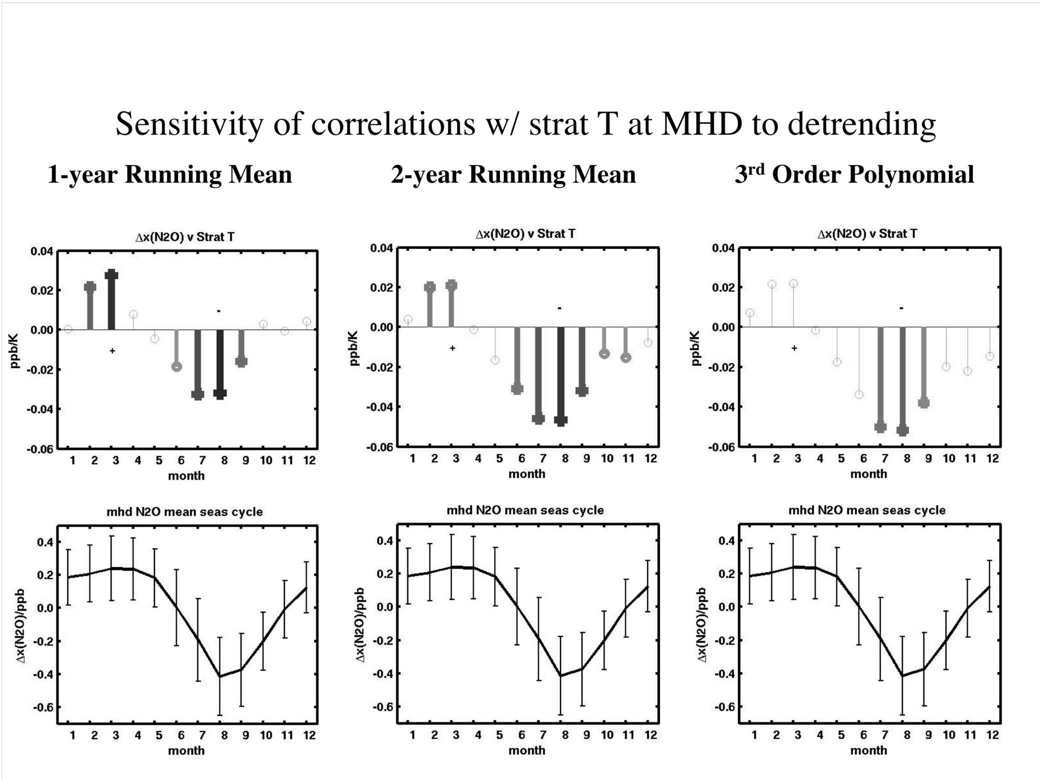

Figure S5. Stem plots summarizing the correlation slopes between detrended AGAGE N_2O monthly means at MHD and polar lower stratospheric temperature (see Figure 6 for details), employing three different detrending filters (see Figure S4). The standard filter used in this paper is the 1-year running mean on the far left.

Figures S6-S8

Below we address the question of whether correlations between polar winter lower stratospheric temperature and detrended tropospheric N_2O seasonal minima, such as those shown in Figures 5-7, can be interpreted as evidence of a stratospheric influence on the tropospheric seasonal cycle, or whether these correlations might arise if tropospheric N_2O variability is driven by weather anomalies, e.g., in convection over continents, that in turn correlate with stratospheric temperatures. Figures S6-S8 provide some model support for the former interpretation while offering no support for the latter interpretation. These figures compare AGAGE observations and model results from *Nevison et al.* [2004], using the Whole Atmosphere Community Climate Model (WACCM), which calculates the stratospheric N_2O sink, but has a fixed, uniform N_2O mixing ratio boundary condition at the surface, and from *Nevison et al.* [2007], using the MATCH:NCEP atmospheric transport model with emissions-based forcing but no stratospheric sink. In brief, the WACCM results in the Northern Hemisphere show regression slope patterns similar to AGAGE data, whereas the MATCH:NCEP results do not.

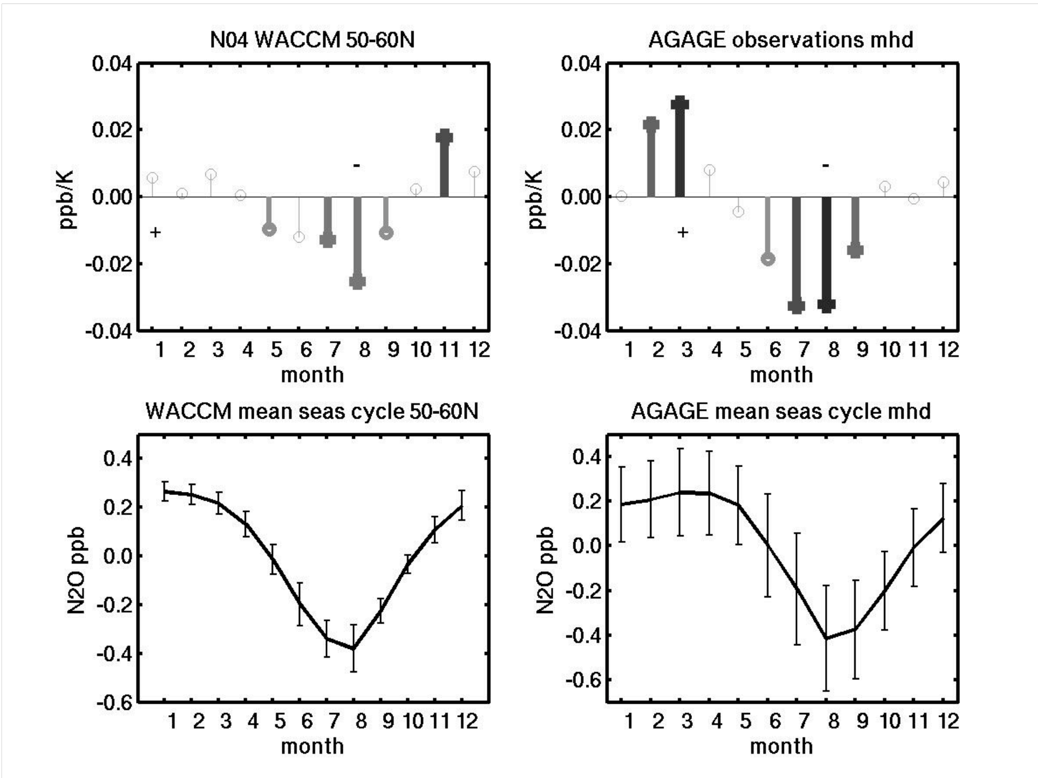

Figure S6. (**Upper Left**) Stem plot summarizing the correlation slopes between detrended 700 mb N_2O monthly means averaged between 50-60ºN from a 13-year simulation of the WACCM model [*Nevison et al*., 2004] and model polar winter lower stratospheric temperature, averaged from January-March at 60-90ºN, 100hPa. Heavy lines indicate statistically significant correlations. The darker the line, the higher the *R* value. The model temperatures and atmospheric dynamics are internally generated and do not necessarily correspond to actual years. Due to the uniform lower boundary condition imposed in WACCM, the seasonal cycles result purely from stratospheric chemistry and stratospheric and/or tropospheric dynamics. Because the restoring influence of the fixed boundary suppresses the seasonal cycles of the model's lowest layers, all tropospheric results are presented at the 700 mb level. (**Lower Left**) mean seasonal cycle in WACCM 700 mb N_2O at 50-60ºN, with error bars showing the standard deviation for each month. **Right-hand panels** are a repeat of Figure 6, which shows the corresponding stem plot and mean seasonal cycle derived for AGAGE N_2O at Mace Head, Ireland.

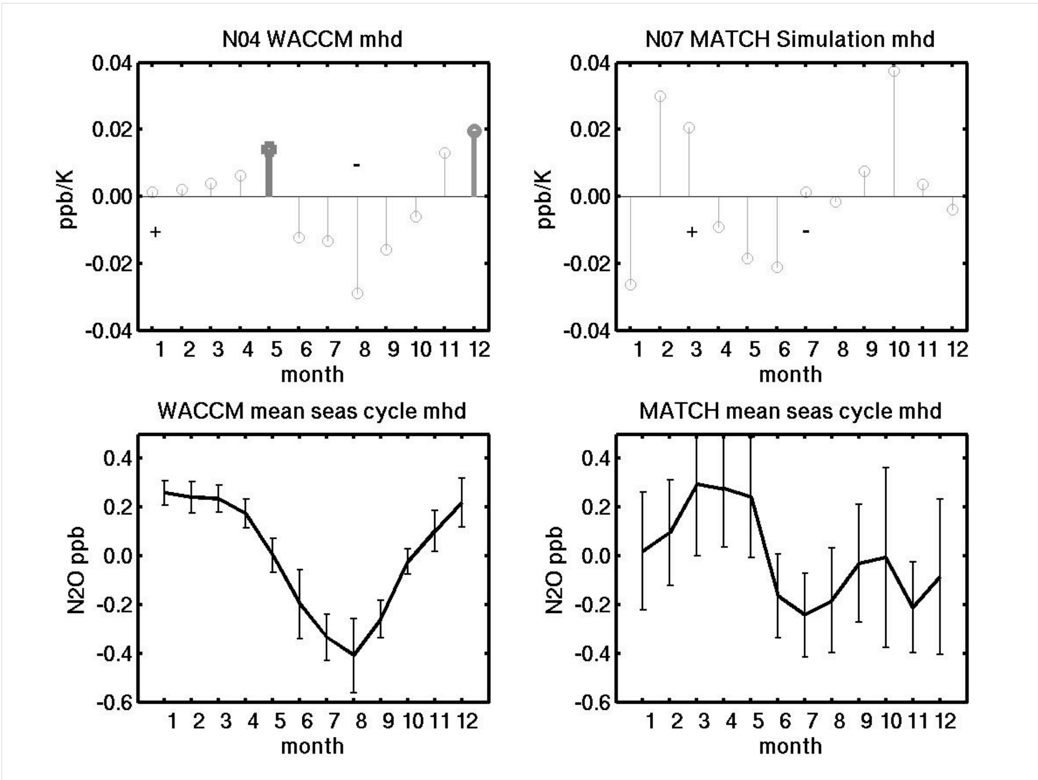

Figure S7. (**Left-hand panels**) Same as Figure S6, but showing WACCM model results sampled at the coordinates of Mace Head, Ireland (53ºN, 10ºW) rather than averaged between 50-60ºN [*Nevison et al*., 2004]. Unlike the 50-60ºN results, the WACCM MHD-specific N_2O correlations with polar winter lower stratospheric temperature are generally not statistically significant, although the same general patterns persist. **Right-hand panels** show the corresponding results of the Nevison et al. [2007] MATCH simulation of N_2O, sampled at Mace Head, suggesting that tropospheric dynamics alone cannot account for the observed seasonal cycle or correlations with stratospheric temperature. The MATCH simulation used monthly-resolved mean surface sources from oceans and natural sources and annually-resolved anthropogenic sources. The simulation was driven with NCAR/NCEP reanalysis data from 1993-2004. The simulation did not include a stratospheric sink. (Note that 50-60ºN zonal mean results are not available from the *Nevison et al.* [2007] simulation.) The regressions in the **Upper Right** panel are performed against NCAR/NCEP polar winter lower stratospheric temperature, the same as for AGAGE data.

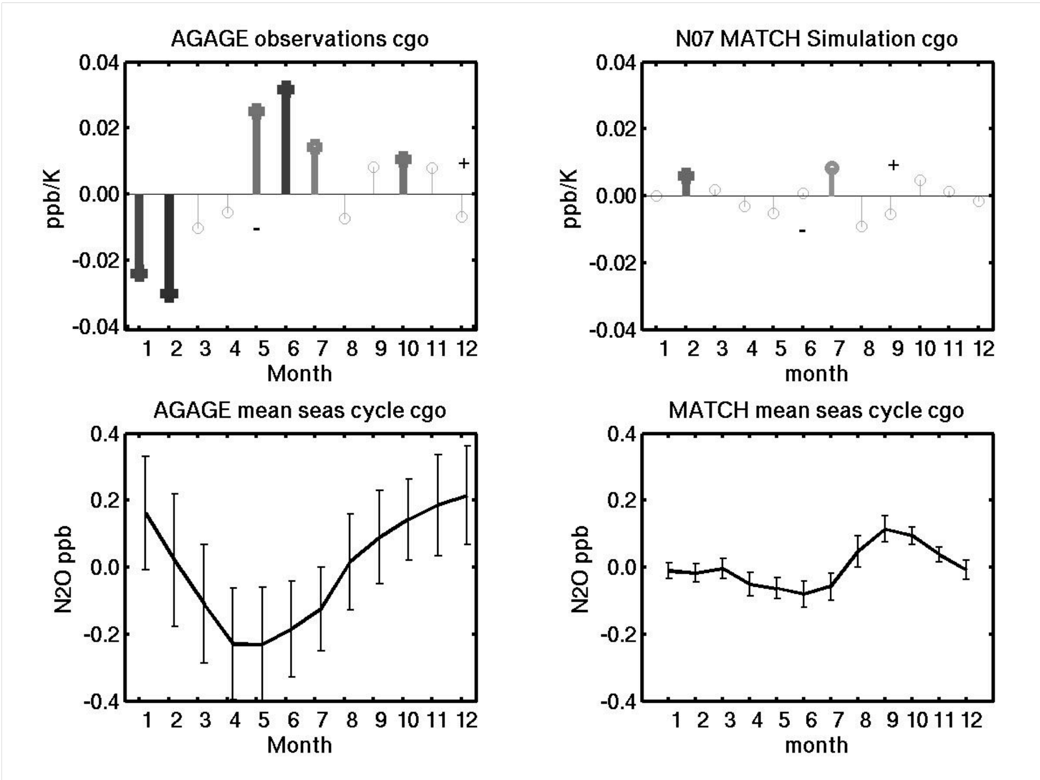

Figure S8. Left-hand panels show a repeat of the AGAGE N_2O results at Cape Grim, Tasmania shown in Figure 10. (**Upper Left**) Stem plots summarizing the linear regression slopes for detrended N_2O monthly means at Cape Grim vs. mean spring southern polar lower stratospheric temperature. **(Lower Left)** mean seasonal cycle for AGAGE N_2O with error bars showing the standard deviation for each month. **Right hand panels** show the corresponding results of the *Nevison et al.* [2007] MATCH simulation of N_2O, sampled at Cape Grim, suggesting that tropospheric dynamics alone cannot account for the observed seasonal cycle or correlations with stratospheric temperature. Note, however, that the WACCM model used in *Nevison et al.* [2004] did not predict a coherent stratospheric influence on the tropospheric seasonal cycle of N_2O in the stratosphere at Cape Grim or elsewhere in the Southern Hemisphere (results not shown).

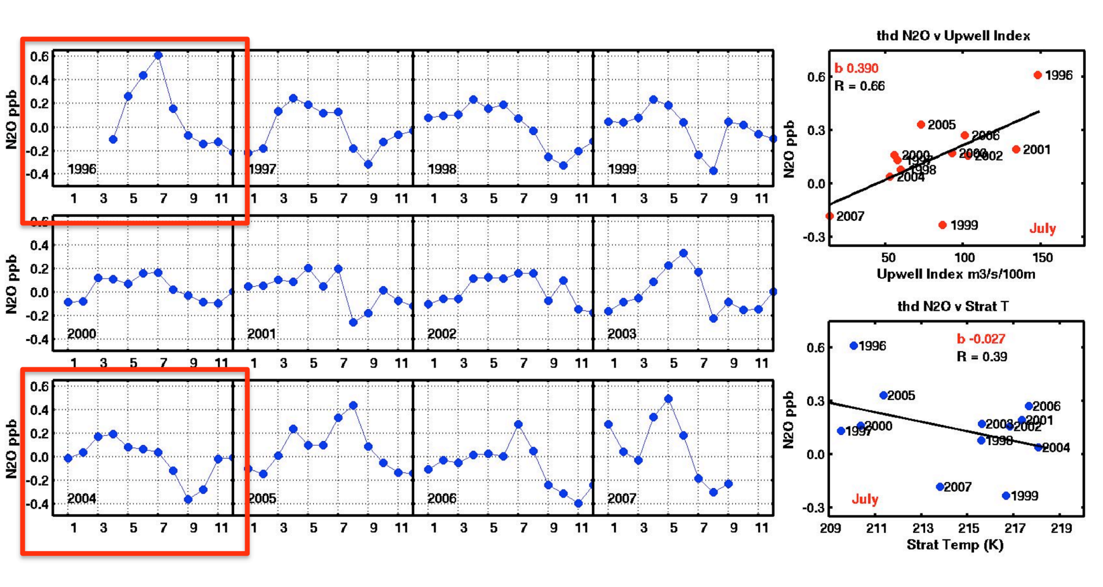

Figure S9. Detrended AGAGE monthly mean N_2O data at Trinidad Head, CA, showing strong interannual variability in the seasonal cycle. 1996 and 2004 are highlighted (red boxes) as years with opposite extremes in the seasonal cycle. Regressions of July detrended N_2O monthly means vs. the NOAA/PFEL coastal upwelling index at 45ºN (upper right) and polar winter lower stratospheric temperature (lower right) show that 1996 was a year of strong July upwelling with a cold stratosphere (corresponding to weak influx), whereas 2004 was a year with weak July upwelling but a warm stratosphere (corresponding to strong influx).

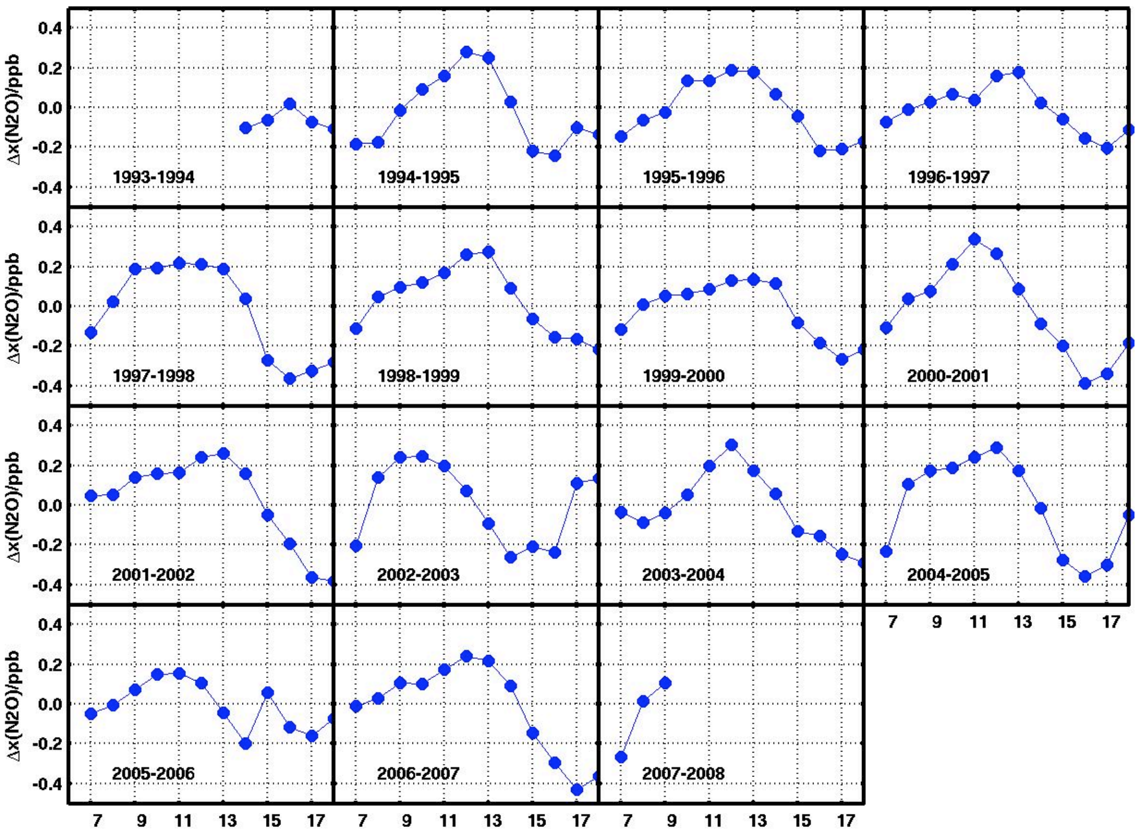

Figure S10. Monthly mean detrended N_2O residuals from the AGAGE network at Cape Grim, Tasmania, repeated from Figure 1b, but plotted from July-June over two calendar years to better identify the maximum in the cycle and the onset of descent into the seasonal minimum.

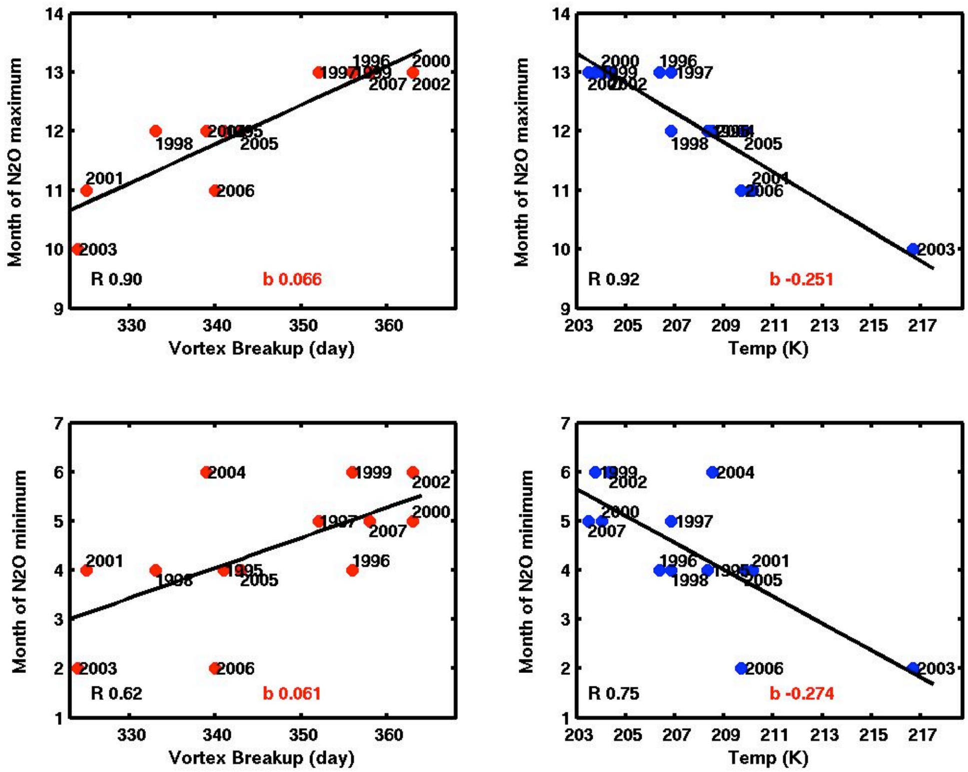

Figure S11. **(Upper Panels)** Month of onset of descent into autumn minimum at Cape Grim, Tasmania (referred to as the month of the N_2O maximum) plotted vs. polar vortex breakup date **(Upper Left)** and mean polar (60-90ºS) spring (Sep-Nov) lower stratospheric (100 hPa) temperature **(Upper Right)**. (**Lower Panels**) Month of the minimum in the monthly mean detrended AGAGE N_2O residuals at Cape Grim, Tasmania (see Figure 1b) plotted vs. polar vortex breakup date **(Lower Left)** and mean polar spring lower stratospheric (100 hPa) temperature **(Lower Right)**. The Pearson's correlation coefficient *R* and the slope *b* of each linear regression are shown. The year labels refer to the second year of the two-calendar-year spans in Figure S10. On the Y-axis, month 13 = January of the second year and month = 14 refers to February of the second year. These plots are conceptually similar to those shown in Figure S3 for Mace Head, Ireland, except that regressions are also shown for the month of the seasonal minimum, which is variable at Cape Grim, in addition to those shown for the month of onset of descent into the minimum , i.e., roughly the month of the maximum. (At Mace Head, the month of the N_2O minimum is more or less constant, occurring always in August.) At other Southern Hemisphere N_2O monitoring sites, including NOAA/CCGG TDF, SYO, HBA and SPO, NOAA/HATS SPO, and CSIRO CGO and SPO, similar correlations are found to those shown in Figure S11. However, these are not always statistically significant and the identification of the seasonal maximum is more ambiguous than for AGAGE CGO, leading to relatively large uncertainty in the regressions.