Interannual variability in tropospheric nitrous oxide

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[1] Observations of tropospheric N_2O mixing ratio show significant variability on interannual timescales (0.2 ppb, 1 standard deviation). We found that interannual variability in N_2O is weakly correlated with that in CFC-12 and SF_6 for the northern extratropics and more strongly correlated for the southern extratropics, suggesting that interannual variability in all these species is influenced by large-scale atmospheric circulation changes and, for SF_6 in particular, interhemispheric transport. N_2O interannual variability was not, however, correlated with polar lower stratospheric temperature, which is used as a proxy for stratosphere-to-troposphere transport in the extratropics. This suggests that stratosphere-to-troposphere transport is not a dominant factor in year-to-year variations in N_2O growth rate. Instead, we found strong correlations of N_2O interannual variability with the Multivariate ENSO Index. The climate variables, precipitation, soil moisture, and temperature were also found to be significantly correlated with N_2O interannual variability, suggesting that climate-driven changes in soil N_2O flux may be important for variations in N_2O growth rate. **Citation:** Thompson, R. L., et al. (2013), Interannual variability in tropospheric nitrous oxide, *Geophys. Res. Lett.*, *40*, 4426–4431, doi:10.1002/grl.50721.

1. Introduction

[2] Nitrous oxide (N_2O) is now the third most important long-lived anthropogenic greenhouse gas. N_2O is also an important ozone-depleting substance (ODS), as it reacts with $O(^1D)$ to produce NO in the stratosphere, and currently the ozone-depleting potential-weighted emissions of N_2O are considered the largest of any ODS [*Ravishankara et al.*, 2009]. The atmospheric mixing ratio of N_2O has been increasing strongly since the preindustrial era largely as the result of human activities, namely the increasing input of reactive nitrogen into ecosystems (predominantly by N fertilizer use), which accelerates denitrification rates and enhances N_2O emissions from soils and coastal waters to the atmosphere [*Syakila and Kroeze*, 2011].

[3] Atmospheric observations of N_2O began in the early 1970s and have revealed a steady quasi-linear increase in concentrations since then [*Prinn et al.*, 1990]. Superimposed on this long-term trend are seasonal and interannual variations. The seasonal variability in N_2O mixing ratio is determined by the combined effect of transport and surface fluxes. Of particular importance is stratosphere-to-troposphere transport (STT), which brings air with a low N_2O mixing ratio from the stratosphere into the troposphere. STT has a maximum in spring in the Northern Hemisphere and is particularly important in defining the seasonal cycle in middle to high latitudes [*Ishijima et al.*, 2010; *Nevison et al.*, 2004; *Nevison et al.*, 2007; *Nevison et al.*, 2011]. The interannual variability in N_2O mixing ratio has only been investigated in a few studies [*Ishijima et al.*, 2009; *Nevison et al.*, 2007; *Nevison et al.*, 2011], which has partly been due to the limited availability of long-term measurements of N_2O precise enough to be used for this purpose. In the late 1990s and early 2000s, measurements of N_2O began at a number of new sites and provide measurements precise enough to be used for such a study. Previous investigations have pointed to an important role of STT in N_2O interannual variability [*Nevison et al.*, 2007; *Nevison et al.*, 2011]. This is based on the observed correlation between anomalies in N_2O minima at middle- to high-latitude sites and anomalies in polar lower stratospheric temperature (PLST), which is used as a proxy for the strength of the Brewer-Dobson circulation and STT [*Huck et al.*, 2005; *Waugh et al.*, 1999].

[4] Here we examine tropospheric N_2O from 1996 to 2009 to better understand the importance of atmospheric transport and surface flux variability on N_2O interannual variability. We exploit a data set encompassing 30 flask and 6 in situ sites (see Table S1 in the supporting information) and utilize observations of two other atmospheric trace gases, CFC-12 (CF_2Cl_2) and SF_6, which are useful tracers for atmospheric transport and have been previously used in investigating seasonal variability in N_2O [*Nevison et al.*, 2007].

2. Observations and Model

[5] Approximately weekly N_2O measurements from discrete air samples (flasks) are used from the NOAA CCGG (Carbon Cycle Greenhouse Gases group) (E. Dlugokencky et al., in preparation, 2013) and the Commonwealth Scientific and Industrial Research Organisation (CSIRO)

Additional supporting information may be found in the online version of this article.

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[*Francey et al*., 2003] global networks. In situ measurements are used from AGAGE (Advanced Global Atmospheric Gases Experiment) [*Prinn et al*., 2000] and NIES (National Institute for Environmental Studies) sites [*Tohjima et al*., 2000] (see Table S1 in the supporting information). For AGAGE sites, the data are available at 40 min intervals while for NIES sites, the data are available as daily averages. Flask and in situ measurements are made using gas chromatographs fitted with electron capture detectors (GC-ECD) and are reported as dry air mole fractions (nmol mol^{-1}, abbreviated ppb). NOAA and CSIRO data are reported on the NOAA-2006A scale [*Hall et al*., 2007], while AGAGE and NIES data are reported on the SIO-1998 and NIES scales, respectively. AGAGE data were adjusted to the NOAA-2006A scale by comparing measurements at sites where AGAGE and NOAA operate in parallel, while NIES data were adjusted based on the results of intercomparisons of standards ($\Delta\chi = 0.6$ ppb, Y. Tohjima, personal communication, 2012). There is some concern that there may be a calibration shift between CCGG N_2O data collected before and after 2001. For this reason, we run our analyses twice, first using all available sites and, second, using a subset of sites ("core" sites), which excludes CCGG sites and sites with large data gaps (>6 months) (see Table S1). CFC-12 and SF_6 measurements are from flasks in the NOAA HATS (Halocarbons and other Atmospheric Trace Species) network and in situ instruments in the AGAGE network. Both measurements are made using GC-ECD and presented as dry air mole fractions (pmol mol^{-1}, abbreviated ppt). CFC-12 measurements are reported monthly on the NOAA-2008 (HATS) and SIO-2005 (AGAGE) scales and SF_6 measurements on the NOAA-2006 (HATS) and SIO-2005 (AGAGE) scales.

[6] Interannual variability (IAV) was calculated for N_2O at each site used in this study by first subtracting the multiannual trend, fitted as a second-order polynomial, and then applying a low-pass Butterworth filter (fourth order) to the residuals to filter seasonal and higher-frequency signals. Two passes of the filter (forward and reverse) were applied to correct for any phase distortion. This method was chosen preferentially over methods that involve fitting a seasonal cycle to the data (e.g., based on harmonic curves) since at many sites the seasonality has small amplitude and/or is irregular. Our definition of IAV is the component of the signal with periodicities longer than 12 months and is closely correlated with the growth rate. The Butterworth filter has been used previously to examine IAV in atmospheric species, e.g., in the studies of *Ishijima et al*. [2009] and *Nakazawa et al*. [1997]. Both methods, however, were tested and gave consistent results. The same method was applied to CFC-12 and SF_6, but for CFC-12 a third-order polynomial was used to fit the multiannual trend.

[7] Atmospheric simulations of N_2O were performed using the Laboratoire de Météorologie Dynamique general circulation model (LMDz, version 4) [*Hourdin et al*., 2006], which includes N_2O photolysis and oxidation reactions in the stratosphere and has an N_2O lifetime of approximately 120 years which is within the range of recent estimates of 131 ± 10 years [*Prather et al*., 2012]. LMDz has a horizontal resolution of 3.75° longitude × 2.5° latitude and 19 hybrid pressure levels up to 3 hPa. Modeled transport was nudged to European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-40 wind fields at 6-hourly intervals. The model was run with climatological monthly N_2O emission estimates comprising natural and agricultural soil fluxes, ocean fluxes, as well as biomass burning and anthropogenic (nonagricultural) emissions (for details see the supporting information).

3. Results and Discussion

[8] IAV in N_2O, CFC-12, and SF_6 is shown for the tropics and the northern and southern extratropics from 1996 to 2009 (Figure 1). The standard deviation of N_2O IAV is approximately 0.2 ppb and is significant compared to the variability across all sites used within each region (for details see Table S2). N_2O IAV is weakly correlated with CFC-12 IAV in the northern extratropics, more strongly correlated in the southern extratropics, and uncorrelated in the tropics (see Table 1). CFC-12, like N_2O, is only lost by photochemical reactions in the stratosphere and has a similar lifetime to N_2O (108 years [*Rigby et al*., 2012]). This, and given that there is little seasonal and interannual variability in CFC-12 emissions, means that it is a pertinent tracer for the influence of STT. The fact that IAV in N_2O and CFC-12 is only weakly correlated in the northern extratropics suggests that STT is not the dominant factor in N_2O variability at these latitudes. IAV in N_2O and SF_6 is also only weakly correlated in the northern extratropics but is more strongly correlated in the tropics and southern extratropics. SF_6 is a very long-lived species (lifetime of 3200 years [*Ravishankara et al*., 1993]) with no loss in the stratosphere, making variations in the tropospheric mixing ratio less sensitive to STT than, e.g., N_2O or CFC-12. In addition, its emissions have little seasonal or interannual variability, making it a useful tracer for tropospheric transport. Emissions of SF_6 are predominantly in the Northern Hemisphere and SF_6 has a strong north-south concentration gradient. Therefore, the correlation of IAV in N_2O with that of SF_6 in the southern extratropics likely results from variations in interhemispheric transport.

[9] Anomalies in N_2O seasonal minima at a number of middle- and high-latitude sites are correlated with anomalies in PLST (used as a proxy for STT) as shown by *Nevison et al*. [2011]. However, we also find substantial variability in the N_2O maxima, which is not correlated with variability in the N_2O minima and cannot be explained by STT alone. Comparing N_2O IAV (which is sensitive to anomalies in both the maxima and minima) in the northern (southern) extratropics with Arctic (Antarctic) PLST, we find no significant correlation (see Table S3). This indicates that STT alone cannot explain the observed IAV in N_2O in the middle to high latitudes. Another test for the influence of STT is to look at the magnitude of this effect. Following *Nevison et al*. [2007], we estimate the change in tropospheric N_2O mixing ratio from variations in STT on interannual timescales. Assuming annually balanced upward and downward air mass fluxes, a N_2O cross-tropopause gradient of 20 ppb, and a net global downward flux of between 4×10^9 and 11.6×10^9 kg s^{-1} (median of 5.9×10^9 kg s^{-1}, $n = 3$) [*Gettelman et al*., 1997; *Schoeberl*, 2004] with a 20% interannual variation (this number is very uncertain [*Schoeberl*, 2004]) results in a range of 0.5–1.4 yr^{-1} interannual variation in the tropospheric mass of N_2O. This equates to 0.11–0.32 ppb IAV in the N_2O tropospheric mixing ratio, of which only the upper limit (i.e., 0.32 ppb) would have about the right magnitude to explain the observed IAV.

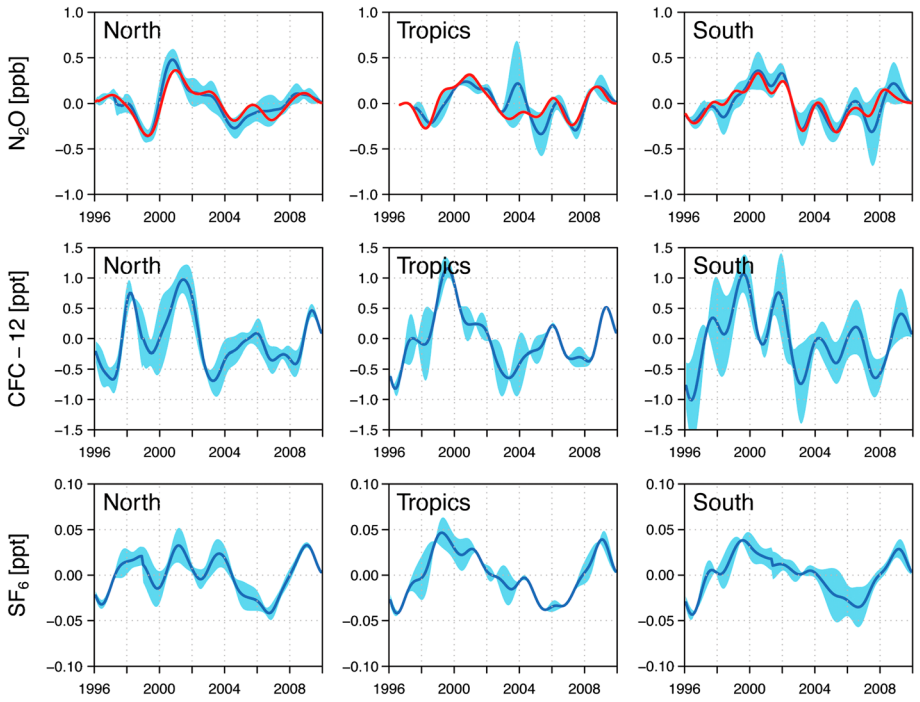

Figure 1. IAV of N_2O, CFC-12, and SF_6 shown for northern (North) and southern (South) extratropical and tropical regions. The solid blue curve is the mean and the shaded blue area is the standard deviation of IAV calculated at all sites. For N_2O, the red line shows the mean value of IAV calculated using the core sites only (see also Table S1).

[10] Examining the latitudinal pattern of N_2O IAV, we find that there is a strong tropical and subtropical signal that is closely in phase across hemispheres and which is negatively correlated with the Multivariate ENSO Index (MEI) (http://www.esrl.noaa.gov/psd/enso/mei/) such that negative N_2O anomalies coincide with El Niño conditions (see Figure 2). A correlation of N_2O IAV with ENSO has been previously detected in long-term N_2O shipboard measurements in the northern and western Pacific [*Ishijima et al.*, 2009] and from in situ measurements at Samoa [*Nevison et al.*, 2007]. We find the strongest correlation with a lag time of between 7 and 9 months in the tropics ($R = -0.61$) and southern extratropics ($R = -0.59$), and between 9 and 11 months in the northern extratropics ($R = -0.67$, see Table 1). In the tropics, CFC-12 IAV (at Samoa) is also correlated with ENSO ($R = -0.41$). Changes in interhemispheric transport, which is affected by ENSO-driven changes in circulation, has been suggested as a mechanism for this correlation and has been observed for other species at Samoa (e.g., methane and methylchloroform) [*Hartley and Black*, 1995; *Prinn et al.*, 1992]. For N_2O the IAV is closely in phase for both hemispheres; however, if changes in interhemispheric transport were mainly responsible for the IAV, then the signal in each hemisphere should be out of phase [*Steele et al.*, 1992]. Also, for species with strong biogenic sources, such as N_2O, this correlation may be partly due to variability in the emissions related to ENSO-driven climate changes. We examine the influence of tropospheric transport and emission variability on N_2O IAV in the following paragraphs.

[11] The transport influence on tropospheric N_2O variability was modeled using LMDz, and time series for each site were extracted from the 4-D simulated N_2O mixing ratios and analyzed in the same way as the observations (see section above). IAV at different sites were then averaged in five geographical regions (see Table S2). Our model (which does not account for IAV in N_2O emissions) captures about 40% of the observed variation in IAV in extratropical regions and none in the tropics (see Figure 3). Simulations using IAV in the emissions (not shown) did not perform better owing to large uncertainties in the emission variability. The model-observation difference for all regions may be partly due to model transport errors, but on the basis of comparisons with CFC-12, for which the agreement is much better ($R = 0.57$ for the northern extratropics and $R = 0.47$ for the

Table 1. Correlation of N_2O IAV With That of the Trace Gases, CFC-12 and SF_6, and With MEI[a]

	Correlation With Tracer Gases		Correlation With MEI	
Region	CFC-12	SF_6	Months of Highest Correlation	Correlation Coefficient
Northern extratropics	0.37	0.35	9–11	−0.67
Tropics	(0.25)	0.51	7–9	−0.61
Southern extratropics	0.68	0.67	7–9	−0.59

[a]All correlations were significant at the 0.05 level, except the correlation of N_2O with CFC-12 in the tropics, which is, therefore, shown in parentheses. The correlation with MEI is shown for the lag times (in months) that gave the highest correlation. The negative correlation means that El Niño events are correlated with negative N_2O anomalies.

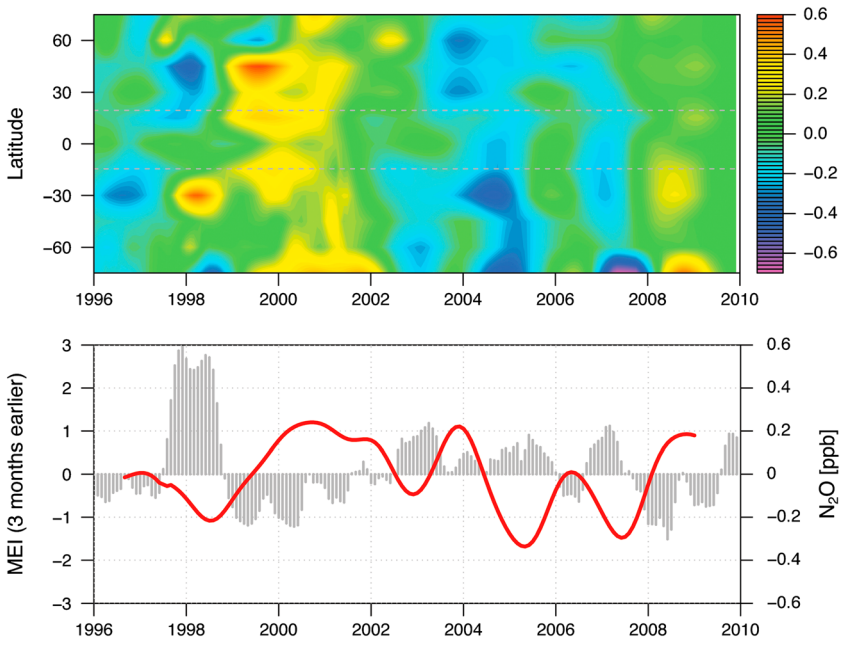

Figure 2. (top) IAV of N_2O (ppb) interpolated with latitude and time. Data were calculated using a bivariate linear interpolation (15° spatial and monthly temporal resolution) of the IAV calculated for each of the 36 sites. The dashed gray lines indicate the latitudinal extent of the stations used in the calculation of the tropical signal. (bottom) MEI shown at monthly resolution with a 3 month lag (gray bars) with the tropical N_2O IAV signal (red line).

tropics, see Table S4), this error is likely to be much less important than the influence of IAV in the emissions. The model-observation comparison suggests that the influence of tropospheric transport is important for N_2O IAV in the extratropics; however, it appears to be less important in the tropics.

[12] Figure 3 shows the IAV in tropospheric N_2O mixing ratios attributed to changes in N_2O emissions (i.e., the observed IAV minus the simulated IAV due to the influence of transport). Denitrification (and to a lesser extent nitrification) in soils is the primary source of N_2O. These processes are known to be very sensitive to soil moisture and temperature, precipitation, soil type, soil pH, and nitrogen substrate availability [*Davidson*, 1993; *Skiba and Smith*, 2000; *Smith et al*., 1998]. We performed an analysis of variance and a correlation analysis of N_2O IAV with the independent variables: soil moisture and temperature, and precipitation (ECMWF ERA-Interim, for details see supporting information), which vary substantially on annual timescales. ERA-Interim soil moisture has been previously evaluated against in situ data (correlation of 0.63 for 2008 to 2010) [*Albergel et al*., 2012]. The analyses were performed on N_2O IAV data with and without the correction for the influence of transport. Due to potential errors in the modeled transport, we refer only to regions where both analyses gave consistent results (see Tables S5 and S6). For Europe, soil temperature and precipitation were positively correlated with N_2O IAV. With the correction for the influence of transport, soil moisture was also positively correlated, and the correlation coefficients increased for all variables. For the tropics, the correlation was calculated with the independent variables for South America, Africa, and Asia, separately. In contrast to Europe, N_2O IAV was negatively correlated with temperature, a result that is consistent with that of *Ishijima et al*. [2009] who found a negative correlation between N_2O IAV and soil temperature in the Northern Hemisphere. N_2O IAV in the tropics was positively correlated with soil moisture (also consistent with *Ishijima et al*. [2009]). The opposing signs of correlation with soil moisture and temperature in the tropics may be a reflection of the fact that anomalies in soil moisture and temperature are often negatively correlated.

[13] A number of extreme climatic events are associated with significant changes in N_2O mixing ratio. The European drought in 2003 produced a strong negative soil moisture anomaly, which coincides with the onset of a significant decrease in N_2O over Europe, which is not observed in either North America or Asia (see Figure S4). In summer 2003, a persistent high-pressure system led to stable conditions and longer air mass residence times over Europe [*Solberg et al*., 2008]. Thus, if no change in surface emissions is assumed, transport alone would result in increased N_2O mixing ratio, while the opposite is observed. Sustained periods of low soil moisture in 2005 and in the first half of 2007 in Asia and South America also coincide with negative N_2O anomalies in the tropical signal, while above average soil water content in 2003 and 2008 in Asia and Africa coincides with positive N_2O anomalies. The years 2005 and 2007 (first half only) are associated with El Niño, which typically brings drier and warmer conditions to South America, Southeast Asia, and South Africa, especially during the Northern Hemisphere winter, whereas 2003 was nearly neutral, and the second half of 2007 and 2008 were La Niña years, which brings generally cooler and wetter conditions to the same regions [*Trenberth et al*., 1998].

[14] Interannual variability in N_2O soil emissions has been previously linked to variations in soil moisture and precipitation, particularly in the tropics [*Potter et al*., 1996; *Werner et al*., 2007], and in soil temperature, particularly in temperate regions [*Potter et al*., 1996]. Low soil moisture limits the production of N_2O via denitrification [*Bouwman*, 1998] and may

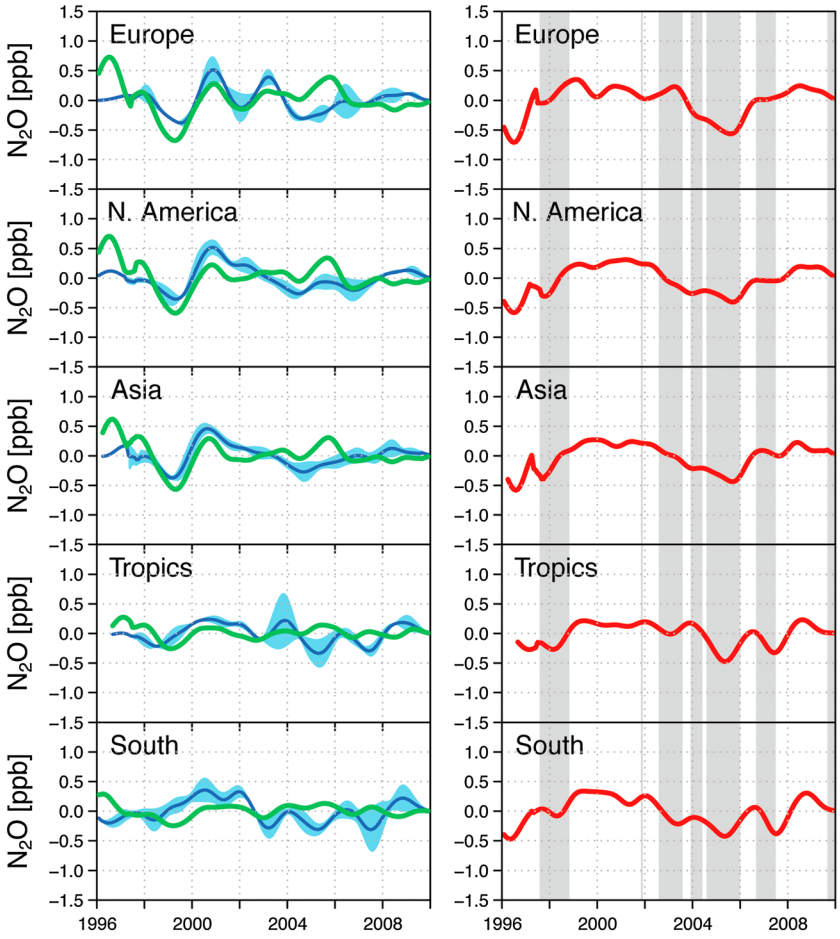

Figure 3. (left) Comparison of observed (blue) and modeled (green) IAV in atmospheric N_2O for different regions. (right) Difference between the observed and modeled IAV, i.e., corrected for the influence of transport. The gray shading indicates El Niño conditions (MEI > 0.2) with a 3 month lag.

reduce the availability of reactive nitrogen in soil by slowing the remineralization of organic matter [*Borken and Matzner*, 2009; *Potter et al*., 1996]. This is opposite to the effect of temperature, which increases the rates of both. The effect of soil temperature and moisture on remineralization rates could be important in areas with little to no N fertilizer input, such as in tropical and subtropical forests, where the background N_2O source is still very large, while their effect on denitrification rates impacts N_2O emissions even in areas that have unlimited reactive nitrogen availability. In this study, we do not investigate correlations between atmospheric N_2O IAV and ocean N_2O fluxes, as from the atmospheric observations alone it is not possible to disentangle fluxes from ocean and land. ENSO influences upwelling in the Tropical Eastern Pacific and, hence, the supply of nutrient- and N_2O-rich water from below the mixed layer [*Behrenfeld et al*., 2006]. *Nevison et al*. [2007] also propose that N_2O IAV in the tropics may be influenced by changes in ocean N_2O flux affected by ENSO. However, the magnitude of the change in N_2O flux, and the corresponding possible change in tropospheric N_2O mixing ratio, is largely unknown.

4. Conclusions

[15] Our analysis shows significant variations in tropospheric N_2O on interannual timescales (0.2 ppb, 1 standard deviation). Comparisons of N_2O, CFC-12, and SF_6 IAV show a weak correlation in the northern extratropics and a stronger correlation in the southern extratropics. The correlation of N_2O with SF_6 IAV in the southern extratropics is likely due to variations in interhemispheric transport, but for CFC-12 other circulation changes may be at play as CFC-12 has only a very weak interhemispheric gradient. We also compared N_2O IAV with PLST (used as a proxy for STT) and found no correlation, indicating that STT is unlikely to be an important factor for year-to-year variations in tropospheric N_2O mixing ratio. On the other hand, N_2O IAV is strongly correlated with MEI, with El Niño being associated with negative N_2O anomalies and vice versa for La Niña. The meteorological parameters, precipitation, soil moisture, and temperature were found to be significant variables for explaining tropospheric N_2O IAV, suggesting that this is modulated by climate-driven changes in soil N_2O emissions. For a more complete understanding of the role climate plays in N_2O IAV, however, more high-quality atmospheric measurements in the tropics would be needed as well as atmospheric inversions to retrieve spatially and temporally resolved N_2O emissions.

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