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Journal
Atmospheric Chemistry and Physics, 23(2)

ISSN
1680-7316

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Publication Date
2023

DOI
10.5194/acp-23-843-2023

Peer reviewed

Atmos. Chem. Phys., 23, 843–849, 2023
https://doi.org/10.5194/acp-23-843-2023

Atmospheric Chemistry and Physics
Open Access
EGU

Observed changes in stratospheric circulation: decreasing lifetime of N_2O, 2005–2021

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Received: 14 September 2022 – Discussion started: 27 September 2022
Revised: 15 December 2022 – Accepted: 16 December 2022 – Published: 18 January 2023

Abstract. Using Aura Microwave Limb Sounder satellite observations of stratospheric nitrous oxide (N_2O), ozone, and temperature from 2005 through 2021, we calculate the atmospheric lifetime of N_2O to be decreasing at a rate of -2.1 ± 1.2 %/decade. This decrease is occurring because the N_2O abundances in the middle tropical stratosphere, where N_2O is photochemically destroyed, are increasing at a faster rate than the bulk N_2O in the lower atmosphere. The cause appears to be a more vigorous stratospheric circulation, which models predict to be a result of climate change. If the observed trends in lifetime and implied emissions continue, then the change in N_2O over the 21st century will be 27 % less than those projected with a fixed lifetime, and the impact on global warming and ozone depletion will be proportionately lessened. Because global warming is caused in part by N_2O, this finding is an example of a negative climate–chemistry feedback.

1 Introduction

Projections of climate change include the acceleration of the stratospheric overturning circulation over the 21st century (Abalos et al., 2021). This three-dimensional circulation, condensed into a two-dimensional framework called the Brewer–Dobson circulation (BDC), brings tropospheric air into the tropical stratosphere where it ascends, being photochemically processed with ultraviolet radiation that increases with altitude, mixes across latitudes, and then descends at mid to high latitudes, re-entering the troposphere around the mid latitudes (Plumb and Mahlman, 1987; Neu and Plumb, 1999; Butchart, 2014). Observational metrics for an enhanced BDC are based on trends in the age of air (AoA) using surrogate gases such as SF_6. While models predict an enhanced BDC, the SF_6 observations indicate an unchanged or decreasing BDC but with large uncertainty (Karpechko et al., 2018; Abalos et al., 2021; WMO, 2022). The comparison of models and measurements of AoA has proven difficult (Fritsch et al., 2020). The search for BDC change with AoA has missed a more obvious and compelling case based on the important greenhouse gas nitrous oxide (N_2O), where recent work has shown agreement in upper stratospheric trends across satellite instruments and a model (Froidevaux et al., 2022). Here we take the Aura Microwave Limb Sounder (MLS) observations of N_2O, ozone (O_3), and temperature (T) from 2005 through 2021 and show that N_2O increases through the middle tropical stratosphere, relatively greater than the rate of tropospheric increases, lead to a shorter atmospheric lifetime, an important consequence of a more vigorous BDC.

Consequences of an enhanced BDC have recently focussed on the increased flux of stratospheric ozone (O_3) into the troposphere with a subsequent increase in tropospheric O_3; see the discussion in Karpechko et al. (2018) and WMO (2022). Enhanced BDC also leads to shorter lifetimes for gases like nitrous oxide (N_2O) and chlorofluorocarbons (CFCs) as greater abundances are pushed higher into the tropical stratosphere, where they experience greater photolytic destruction. Two decades ago this mechanism was proposed by Butchart and Scaife (2001) for the CFCs based on dynamical diagnostics of the BDC from a climate model. Now we have observed and quantified this effect for a major greenhouse gas. Using the simultaneous MLS vertical

profiles of N_2O, O_3, and temperature, we calculate the total loss of N_2O in the column, needing only the solar spectrum and absorption cross sections, as in Prather et al. (2015, hence P2015). We find that the stratospheric N_2O loss rate is increasing more quickly than the observed trend in surface N_2O, and hence the N_2O lifetime is declining. For N_2O, a major greenhouse gas, it means that the climate impact of its anthropogenic emissions will be reduced over this century, and for CFCs, where we expect parallel results, it means that these now-banned ozone-depleting gases will be cleaned out of the atmosphere more quickly, as shown by Butchart and Scaife (2001).

2 Measurements and methods

The Aura MLS observations of N_2O, O_3, and T are taken from the Goddard Earth Sciences Data and Information Services Center in the form of V5 Level-3 monthly binned profiles from August 2004 through December 2021 (Lambert et al., 2021; Schwartz et al., 2021a, b). All data are averaged over the 72 longitude bins. There are 43 latitude profiles every 4° (84° S to 84° N). Pressure levels range from 100 to 0.001 hPa with logarithmic spacing, and the resolution depends on the MLS quantity. The very few negative O_3 values (0.04 % of data at 0.01 hPa) occur at the minimum in the O_3 profile and are replaced with the mean value of 0.2 ppm, with negligible impact on photolysis rates. The negative N_2O values are slightly more frequent (about 0.1 % of data at 32 and 22 hPa but 1.6 % at 0.5 hPa, where the mean value is 2.3 ppb). We do not wish to inflate the N_2O loss by replacing negative values with large positive ones, so we opt for the safest choice and just replace negative N_2O values with 1 ppb, again with negligible impact on N_2O loss. The N_2O vertical grid between 146 and 1 hPa (six evenly spaced log(pressure) levels per decade) is coarser than that of O_3 and T (both use 12 intervals per decade). The missing N_2O levels in the O_3–T grid are calculated as the square root of the product of the values on the adjacent levels. For pressures <0.4 hPa, where MLS V5 N_2O data are not reported, we set the abundance to 0 ppb. Our consolidated MLS dataset then covers 43 latitudes (84° S to 84° N), 37 pressure levels (100–0.001 hPa), and 209 months (August 2004 to December 2021).

The observational data consist of 8987 independent column atmospheres. The top level (0.001 hPa) is high enough that only a single layer of thickness 0.001 hPa with the same properties as the top layer is added to complete the atmosphere for the radiative calculation. Below 100 hPa, we add six layers with typical tropospheric values (320 ppt); however, these layers have a negligible impact on N_2O loss because very little of the critical ultraviolet radiation reaches below 100 hPa. Each latitude profile is weighted by the area of the Earth ±2° latitude on either side except for latitudes 1 (90–82° S) and 43 (82–90° N). For each column we calculate the profile of N_2O loss from photolysis (J-N_2O) and reaction with the electronically excited atomic oxygen $O(^1D)$ from the three reactions.

$$N_2O + h\nu \rightarrow N_2 + O \quad \text{(R1)}$$

$$N_2O + O(^1D) \rightarrow N_2 + O_2 \quad \text{(R2)}$$

$$N_2O + O(^1D) \rightarrow NO + NO \quad \text{(R3)}$$

The $O(^1D)$ is calculated from the O_3–T profiles assuming production equals loss.

$$O_3 + h\nu \rightarrow O(^1D) + O_2 \quad \text{(R4)}$$

$$O(^1D) + N_2 \rightarrow O(^3P) + N_2 \quad \text{(R5)}$$

$$O(^1D) + O_2 \rightarrow O(^3P) + O_2 \quad \text{(R6)}$$

The radiative transfer calculation is the same as in Prather et al. (2015) and includes the solar declination for the middle of each month, the Sun–Earth distance, but not the solar cycle (a mean solar flux is used).

Vertically, losses are weighted linearly in pressure (mass), and across latitude, they are weighted by the area of each latitude belt. Annually integrated budgets include the number of days in each month, but leap years are treated as 365 d years, and thus our annual losses are biased low by 0.07 %. Lifetimes are budgetary timescales and calculated as a 12-month burden (TgN) divided by the 12-month loss ($TgN\,yr^{-1}$). The burden is calculated from monthly-mean, globally-averaged NOAA marine surface measurements (Dlugokencky, 2022) and uses the conversion factor of $4.78\,TgN\,ppb^{-1}$ (Prather et al., 2012). Given that the gridding and source files have changed since P2015 (which used the 5° latitude GOZCARDS data), we compared the global monthly loss rates for the overlap period with P2015 (August 2004–December 2011): the mean difference of 0.2 % and root mean square (rms) difference of 0.6 % show that both calculations are essentially identical. Uncertainty in the absolute N_2O lifetime is estimated at ±8 % (1-sigma), based primarily on the absolute calibration of MLS N_2O and the chemical kinetics uncertainty; see Sect. 4 of P2015.

Uncertainties in trends here are calculated from a linear regression of the 12-month running averages of the monthly means as reported or calculated here. We select the 68 % (1-sigma) range, because we are looking for "likely" connections (68 % confidence) rather than "extremely likely" ones (95 %). For example, the linear regression trend in N_2O loss uses the running averages and has 198 monthly points. The uncertainty is scaled up assuming there are only 17 yearly data points. As a check we recalculate the regression fit with the unsmoothed monthly data and get the same uncertainty.

3 Results

The primary calculation here is the annual N_2O loss ($TgN\,yr^{-1}$) shown in Fig. 1a. It was surprising in that such a clear 17-year increase is apparent. Most of the

interannual variability in this 12-month running mean time series is associated with the quasi-biennial oscillation (see Ruiz et al., 2021). The linear regression gives a trend of $+5.0 \pm 0.7$ %/decade. This is larger than the $+2.9 \pm 0.02$ %/decade trend in the burden (Fig. 1b) and results in an N_2O budgetary lifetime trend of -2.1 ± 0.7 %/decade (Fig. 1c). Our best estimate for the N_2O lifetime over the past decade (117 years) is still close to that in P2015, which included other model calculations. Most important for the N_2O budget is the average loss itself, namely, 13.43 TgN yr^{-1} with an interannual standard deviation of 3 % and a min-to-max range of 7 %. The primary source of stratospheric odd-nitrogen species (e.g., NO, NO_2, HNO_3) is production of NO in Reaction (R3), and we calculate the production of NO (Figure 1d), which (like the N_2O loss) also shows a positive trend of $+3.9 \pm 0.6$ %/decade.

The immediate cause of the lifetime trends (i.e., enhanced N_2O loss) can be an increase in the photochemical loss frequency or in the abundance of N_2O, or both. The critical zone for loss in the vertical is 3 to 30 hPa (80 % of total loss), and in latitude it is 30° S to 30° N (75 %); see P2015. Thus we focus on the tropical middle stratosphere. Photolysis (Reaction R1) dominates loss here (by more than 90 %), and the monthly mean J-N_2O at the Equator (Fig. 1e) shows only a consistent decline across the critical region from -0.5 ± 0.1 %/decade at the top of the region (3 hPa) to -1.3 ± 0.3 %/decade at altitudes below 10 hPa. This change by itself would reduce the loss.

The declining J-N_2O is primarily due to the recovery of overhead ozone during this period from reduced chlorine-catalyzed depletion as the CFCs and other chlorinated gases have declined following the Montreal Protocol agreement that regulated these source gases (Bernath et al., 2020; Froidevaux et al., 2022, and references therein). Observations show continued increases in upper tropical stratospheric ozone for 2000–2020 (Godin-Beekmann et al., 2022) from a range of satellite measurements, including the MLS O_3 used here that is driving the change in J-N_2O. For the upper stratospheric tropics (1–10 hPa), the generally observed O_3 trend of order $+1.0$ %/decade to $+1.6$ %/decade is consistent with the vertical pattern and magnitude of the J-N_2O trend. Another source of declining J-N_2O is the reduction in photolysis cross sections from lower temperatures. We calculate that the relative change in J in this altitude region is $+0.4$ % K^{-1}, and combined with the observed temperature trend of about -0.5 K/decade (Maycock et al., 2018), we estimate that the trend in J from temperature-dependent cross sections is -0.2 %/decade and thus a small contribution to the total. The overall trend in J of about -1 %/decade reduces the N_2O loss and makes the disagreement between the burden and loss trends even greater.

Given that N_2O loss is increasing faster than the global burden by $+2.1$ %/decade, we expect that the N_2O abundances in the critical zone are likewise increasing more quickly. Because of the reduction in photolysis rates, we expect the abundance to be increasing at about $+6$ %/decade. Can we see this in the MLS N_2O data? The monthly mean 30° S–30° N N_2O abundances are shown in Fig. 1f. The trends are clear but vary over altitude. At the bottom of the critical zone (32 hPa) the trend is negative, -3.4 ± 0.5 %/decade, and is probably due to a residual negative drift of the V5 MLS data in the lower stratosphere after 2010 as shown in Fig. 16 of Livesey et al. (2021). It is unlikely to be real because suppression of vertical transport of N_2O below 30 hPa can hardly lead to increases in N_2O above and, moreover, the ACE-FTS data show positive trends at these pressures following the analysis of Froidevaux et al. (2022). The trend increases rapidly, reaching $+5.5 \pm 1.2$ %/decade in the central zone (10 hPa) and $+12.0 \pm 2.3$ %/decade near the top (4.6 hPa). These increases are used directly in our calculation and average to the $+6$ %/decade needed to explain the trend in total N_2O loss.

Are these increases in N_2O real or an artifact of a known calibration drift in the MLS 190 GHz observations, affecting mainly water vapor and N_2O? The MLS V5 dataset benefits from an extensive effort to remove/reduce drifts and verify the MLS H_2O and N_2O against the overlapping ACE-FTS satellite measurements, which are not believed to be drifting (Livesey et al., 2021; Froidevaux et al., 2022). The key comparison is Fig. 16 of Livesey et al. (2021). In the tropics (20° S–20° N) there could be a small positive drift in MLS V5 N_2O versus ACE-FTS N_2O for the period 2005–2010 in the range 3–30 hPa, but for the later period 2010–2019, there is no drift, or even a negative drift at 30 hPa and below. This change with altitude may explain the small or negative trend in N_2O below 15 hPa. It is possible that N_2O is increasing throughout the tropical stratosphere if the negative trend at 32 hPa is due to instrumental drift. Parallel analysis of the MLS V4 N_2O (not shown) shows a similar increase in the trend from small negative trends at 32 hPa to large positive ones at 3 hPa (not shown). At the upper end of our range (2.2–6.8 hPa), Froidevaux et al. (2022, their Fig. 12) find similar trends in N_2O to ours (~ 13 %/decade) from MLS, ACE-FTS, and a model. Their analysis included a much wider latitude range (50° S–50° N), but because N_2O abundances fall off rapidly outside of the tropical ascent region (see Fig. 3 of Prather et al., 2015), the tropics should dominate the mean value and its trend. Thus, any calibration drift that impacts the lifetime (i.e., occurring in the critical region 3 to 30 hPa and 30° S to 30° N) is negligible compared to the increasing trend in N_2O loss ($+5.0$ %/decade). If the drift were slightly negative, correction would further increase this trend.

We believe that the solar cycle has had little impact on N_2O lifetime variability over the Aura MLS record here. The period 2005–2011 following Cycle 23 showed low activity (smoothed monthly sunspot number <50), the peak of Cycle 24 (2011–2015) was among the lowest in the last 100 years, and Cycle 25 began in 2020 but activity remained low through 2021. We conclude that the impact of the solar cycle on these observations was much less than the min-to-max de-

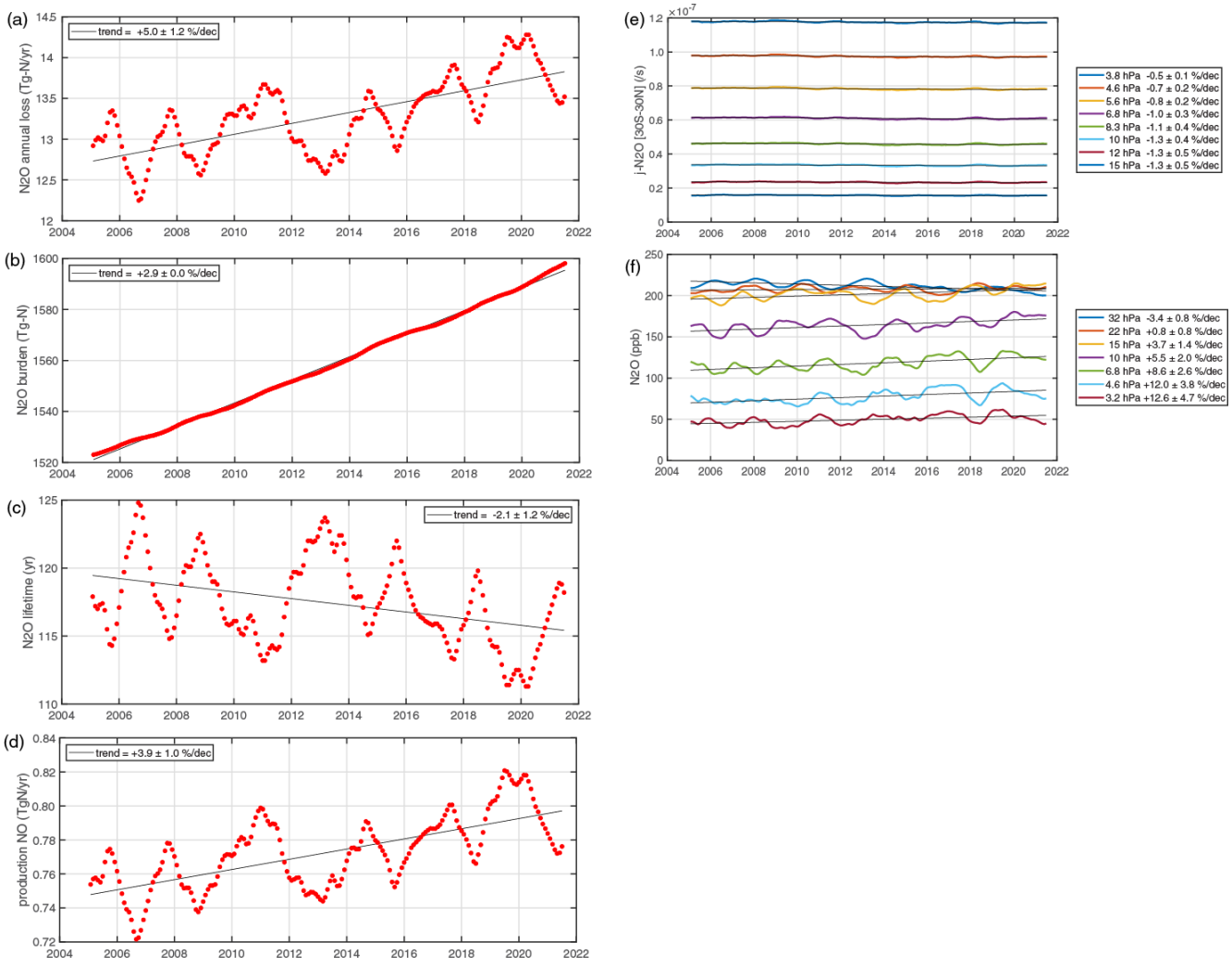

Figure 1. **(a)** N_2O loss rate (TgN yr^{-1}) as monthly values of a 12-month running mean. The 198 red points begin with the 12-month average of August 2004 through July 2005 plotted as 1 February 2005 with the final point being 1 July 2021 (all of 2021). The thin black trend line shows the slope and uncertainty in the legend; see the text. **(b)** N_2O global burden (TgN) based on marine surface observations. **(c)** N_2O lifetime derived from the burden divided by loss rate. Production of NO (TgN yr^{-1}) from N_2O loss. **(e)** Stratospheric photolysis rates, J-N_2O (/s), at the standard MLS V5 *T* pressure levels (see legend) and averaged over 30° S–30° N. **(f)** N_2O abundance (ppb) at the standard V5 N_2O pressure levels (see legend) and averaged over 30° S–30° N.

crease in N_2O lifetime of 4 %–7 % (see P2015) and probably less than 2 %–3 %. Moreover, the symmetry of this low solar activity over 2005–2021 is unlikely to affect trends.

4 Implications

The recent WMO Ozone Assessments (Karpechko et al., 2018; WMO, 2022) concluded that disagreement remains regarding the direction of the BDC trend between the chemistry–climate model simulations (increasing rates) and the satellite observations of AoA tracers (decreasing or uncertain). We present clear observational evidence supporting the models of what is likely a climate-change-driven increase in the net BDC using a major trace gas. N_2O abundances in the tropical upper stratosphere are increasing at rates much faster than tropospheric abundances, but this pattern cannot distinguish between more rapid ascent or reduced mixing with extra-tropical latitudes. The latter would imply a slower growth rate for extra-tropical N_2O, but this is not seen. If we perform a similar trend analysis to that shown in Fig. 1f for the extra-tropics (i.e., 30–58° N and 30–58° S), then we find an almost identical pattern to that in the tropics: increasing at 7 %/decade–8 %/decade versus 6 at 10 hPa and at 14 %/decade–18 %/decade versus 13 at 3.2 hPa; see Supplement Fig. S1 and Table S6. The obvious explanation is an increase in the meridional mean ascent rate in the tropics with little or no change in mixing across the barrier between the tropics and extra-tropics. The use of an integrated quan-

tity like the N_2O lifetime provides robust averaging over the large seasonal and interannual variability of this gas in the middle and upper stratosphere. We expect these results to hold for other gases with mid-stratosphere photolytic sinks, such as CFC-12.

Viewing the changing BDC in terms of lifetimes gives a different perspective of the potential consequences of climate change. If this rate of change continues over the 21st century, then the lifetimes of N_2O and CFC-12 might drop by 20 %. Thus CFCs will be cleared out of the atmosphere earlier than currently projected (e.g., Butchart and Scaife, 2001). The climate impact of N_2O emissions will drop because their decay time will fall from 110 to 90 years. Note that the decay time of a pulse based on a lifetime of 117 years is reduced by a factor of 0.94 due to chemical feedbacks (Prather, 1998; Prather et al., 2015). For example, if we extrapolate the observed growth in N_2O burden over the last 2 decades (+2.9 %/decade), we reach 419 ppb by 2100, a value between RCP6.0 and RCP8.5. If we use a constant lifetime of 120 years to derive annual emissions and then re-project future N_2O using a declining lifetime (−2.1 %/decade), this value drops to 391 ppb (see Fig. S2). Over the 21st century, the N_2O increase drops by 27 % from 103 to 75 ppb, corresponding to a drop in effective radiative forcing of 0.09 W m^{-2}, equivalent to about 6.6 ppm CO_2 (Forster et al., 2021). Such differences are substantial when trying to tune our mitigation strategies to achieve climate change goals.

Overall, we will see an accelerated removal of the long-lived trace gases. Chipperfield et al. (2014) analyzed the change in CFC and N_2O atmospheric lifetimes with climate change (year 2100 versus 2000). For the five chemistry–climate models that calculated the N_2O lifetime, results were ambiguous: two increased, one decreased, and two were unchanged, all with absolute changes of less than 0.5 %/decade. These results are not necessarily inconsistent with the projections here because the modeled scenarios included other chemical changes in the stratosphere and adopted a middling climate scenario (RCP 4.5). Chipperfield et al. (2014) also found that AoA metrics were a poor predictor of N_2O lifetime (see their Figs. 8–9). Thus, chemistry–climate model intercomparison projects should encourage the calculation of N_2O and CFC lifetimes over the 21st century as a major diagnostic of changing BDC, although accompanying changes in O_3 and T must also be diagnosed to evaluate the photochemically driven changes in lifetime.

There is an additional oddity in this analysis. We find that the production of NO (+3.9 %/decade) is increasing more quickly than the burden (+2.9 %/decade) but not as quickly as N_2O loss (+5.0 %/decade), although the uncertainties overlap. This is expected because NO production peaks lower in the stratosphere and the rapid increase in loss above 10 hPa (∼ 32 km) produces proportionally much less NO (see P2015, Fig. 1). Reduced NO production relative to N_2O loss was also a theme in Nevison et al. (1999), who suggested that such changes might occur and be detectable in the observed ratio of NOy (NO and its products) to N_2O in the lowermost stratosphere. Our results are consistent with the analysis of Froidevaux et al. (2022), where the abundance of NO and NO_2 in the upper stratosphere is increasing at a rate much lower than that of N_2O (approximately 2 versus 12 %/decade). The abundance of NO in the tropical upper stratosphere, however, is a balance between production, vertical transport, and photochemical loss above 40 km, and we cannot estimate it from the production alone.

In parallel with our MLS N_2O data analysis, S. E. Strahan led a full chemistry-transport modeling study of the MLS period using the MERRA-2 wind fields and focusing on the chemistry of N_2O, NO_y, and O_3. Their results support our analysis and show that more rapid vertical ascent in the tropical upper stratosphere is the cause of enhanced N_2O and N_2O loss. This enhanced transport of NO led to its accumulation in the Arctic upper stratosphere, with enhanced O_3 loss there. The modeled changes were shown to be driven by circulation changes, and they were matched by available satellite observations. That paper has now appeared (Strahan et al., 2022).

From the results here and in P2015, we assemble a budget for N_2O from pre-industrial to the last 2 decades (Table S7). Assuming that the pre-industrial stratospheric loss (10.5 TgN yr^{-1}) is natural and has remained constant, then the anthropogenic emission for the 2000s decade is 6.0 TgN yr^{-1} and that for the 2010s decade is 7.6 TgN yr^{-1}. This 27 %/decade increase in anthropogenic emissions comes primarily from the increasing N_2O atmospheric burden, with 4 %/decade of the total coming from the decrease in N_2O lifetime derived here. Such growth in emissions shows how hard it will be to control them (Davidson, 2012) and identifies N_2O as a looming threat to climate stabilization.

Data availability. The raw data used here were downloaded from the sites and at the dates specified (https://gml.noaa.gov/webdata/ccgg/trends/n2o/n2o_mm_gl.txt, Dlugokencky, 2022; https://disc.gsfc.nasa.gov/datasets/ML3MBN2O_005/summary, Lambert et al., 2021; https://disc.gsfc.nasa.gov/datasets/ML3MBT_005/summary, Schwartz et al., 2021a; https://disc.gsfc.nasa.gov/datasets/ML3MBO3_005/summary, Schwartz et al., 2021b). The time series calculated here and shown in the figures are included in Tables S1–S5 and S8 in the Supplement.

Supplement. The supplement related to this article is available online at: https://doi.org/10.5194/acp-23-843-2023-supplement.

Author contributions. MJP designed the research and performed the data analysis. LF and NJL analyzed the accuracy of the datasets. MJP wrote the paper. LF and NJL reviewed and edited the paper.

Competing interests. The contact author has declared that none of the authors has any competing interests.

Acknowledgements. We thank Jessica Neu for helpful framing discussions and other MLS team members (Alyn Lambert, William Read, and Ryan Fuller) who contributed to the MLS V4 and V5 datasets used here (O_3, T, and especially N_2O). We also thank the two anonymous *ACP* reviewers whose constructive reviews improved the manuscript.

Financial support. Research at the University of California Irvine was supported by grants from the National Aeronautics and Space Administration's Atmospheric Chemistry Modeling and Analysis Program (ACMAP; grant no. 80NSSC21K1454) and the National Science Foundation's Atmospheric Chemistry Program (grant no. AGS-2135749). Work at the Jet Propulsion Laboratory, California Institute of Technology, was performed under contract with NASA (80NM0018D0004).

Review statement. This paper was edited by Susannah Burrows and reviewed by two anonymous referees.

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Atmospheric Chemistry and Physics
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Supplement of

Observed changes in stratospheric circulation: decreasing lifetime of N_2O, 2005–2021

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This Supplement contains data tables of the time series plotted in the paper. It also includes the time series and figures for the N_2O stratospheric abundances (f-N_2O) in northern and southern mid-latitude stratosphere that were discussed but not plotted or shown un the main text.

Note that Tables S1-S5 contain 12-month running mean data (from 2005.08 to 2021.5), while Table S8 contains the MLS-calculated monthly N_2O loss data (2004.63 to 2021.96).

Contents:

Table S1.

year	loss (TgN/y)	burden (TgN)	lifetime (y)	prod NO (TgN/y)
	fig 1A	**fig 1B**	**fig 1C**	**fig 1D**
2005.08	12.92	1523.0	117.90	0.7538
2005.17	12.99	1523.3	117.20	0.7570
2005.25	13.02	1523.5	117.00	0.7578
2005.33	13.00	1523.9	117.30	0.7563
2005.42	12.98	1524.2	117.40	0.7551
2005.50	13.04	1524.5	116.90	0.7586
2005.58	13.20	1524.8	115.50	0.7670
2005.67	13.33	1525.2	114.40	0.7734
2005.75	13.35	1525.5	114.30	0.7746
2005.83	13.29	1525.9	114.80	0.7720
2005.92	13.17	1526.3	115.90	0.7670
2006.00	13.04	1526.7	117.10	0.7616
2006.08	12.91	1527.1	118.30	0.7555
2006.17	12.76	1527.5	119.70	0.7484
2006.25	12.65	1527.9	120.80	0.7430
2006.33	12.58	1528.2	121.50	0.7392
2006.42	12.54	1528.5	121.90	0.7375
2006.50	12.47	1528.8	122.60	0.7330
2006.58	12.34	1529.1	123.90	0.7261
2006.67	12.25	1529.3	124.80	0.7216
2006.75	12.27	1529.5	124.60	0.7225
2006.83	12.36	1529.7	123.70	0.7269
2006.92	12.50	1529.9	122.40	0.7330
2007.00	12.62	1530.2	121.20	0.7385
2007.08	12.75	1530.4	120.00	0.7441
2007.17	12.88	1530.6	118.80	0.7505
2007.25	12.97	1530.9	118.00	0.7552
2007.33	13.03	1531.2	117.50	0.7588
2007.42	13.05	1531.5	117.30	0.7603
2007.50	13.09	1531.8	117.00	0.7625
2007.58	13.16	1532.2	116.40	0.7669
2007.67	13.28	1532.6	115.40	0.7736
2007.75	13.36	1533.0	114.80	0.7781
2007.83	13.35	1533.5	114.90	0.7779
2007.92	13.27	1534.0	115.60	0.7743
2008.00	13.17	1534.5	116.50	0.7703
2008.08	13.06	1534.9	117.60	0.7652
2008.17	12.93	1535.4	118.80	0.7587
2008.25	12.83	1535.8	119.70	0.7537
2008.33	12.79	1536.2	120.20	0.7516
2008.42	12.79	1536.5	120.10	0.7518
2008.50	12.79	1536.9	120.10	0.7517
2008.58	12.75	1537.2	120.60	0.7490
2008.67	12.68	1537.6	121.30	0.7443
2008.75	12.58	1537.9	122.20	0.7390
2008.83	12.56	1538.2	122.50	0.7376
2008.92	12.62	1538.5	121.90	0.7400
2009.00	12.71	1538.6	121.10	0.7437
2009.08	12.80	1538.9	120.20	0.7476
2009.17	12.88	1539.1	119.50	0.7509

2009.25		12.93		1539.4		119.00	0.7532
2009.33		12.94		1539.8		119.00	0.7532
2009.42		12.96		1540.1		118.80	0.7538
2009.50		13.06		1540.4		118.00	0.7584
2009.58		13.17		1540.7		117.00	0.7642
2009.67		13.25		1541.0		116.30	0.7684
2009.75		13.30		1541.2		115.90	0.7707
2009.83		13.31		1541.6		115.80	0.7715
2009.92		13.31		1542.0		115.90	0.7713
2010.00		13.29		1542.4		116.10	0.7707
2010.08		13.29		1542.8		116.10	0.7718
2010.17		13.36		1543.2		115.50	0.7763
2010.25		13.40		1543.6		115.20	0.7798
2010.33		13.41		1544.0		115.10	0.7816
2010.42		13.37		1544.4		115.60	0.7806
2010.50		13.28		1544.8		116.30	0.7776
2010.58		13.26		1545.3		116.50	0.7780
2010.67		13.31		1545.8		116.10	0.7817
2010.75		13.43		1546.2		115.20	0.7887
2010.83		13.53		1546.7		114.40	0.7932
2010.92		13.62		1547.1		113.60	0.7971
2011.00		13.67		1547.6		113.20	0.7989
2011.08		13.67		1548.0		113.20	0.7981
2011.17		13.62		1548.4		113.70	0.7943
2011.25		13.57		1548.8		114.10	0.7911
2011.33		13.55		1549.1		114.30	0.7890
2011.42		13.58		1549.4		114.10	0.7896
2011.50		13.60		1549.8		114.00	0.7892
2011.58		13.57		1550.2		114.20	0.7867
2011.67		13.47		1550.5		115.10	0.7800
2011.75		13.32		1550.9		116.40	0.7720
2011.83		13.21		1551.2		117.40	0.7669
2011.92		13.09		1551.5		118.50	0.7615
2012.00		13.01		1551.8		119.30	0.7578
2012.08		12.97		1552.1		119.70	0.7562
2012.17		12.97		1552.5		119.70	0.7569
2012.25		12.98		1552.9		119.60	0.7577
2012.33		12.98		1553.2		119.60	0.7578
2012.42		12.92		1553.5		120.20	0.7551
2012.50		12.81		1553.8		121.30	0.7502
2012.58		12.74		1554.1		122.00	0.7476
2012.67		12.74		1554.3		122.00	0.7483
2012.75		12.76		1554.6		121.90	0.7493
2012.83		12.75		1554.9		122.00	0.7492
2012.92		12.71		1555.3		122.30	0.7482
2013.00		12.66		1555.7		122.90	0.7466
2013.08		12.61		1556.1		123.40	0.7450
2013.17		12.58		1556.5		123.70	0.7440
2013.25		12.61		1556.9		123.40	0.7462
2013.33		12.69		1557.2		122.70	0.7509
2013.42		12.79		1557.6		121.80	0.7559
2013.50		12.85		1558.0		121.20	0.7590
2013.58		12.80		1558.4		121.70	0.7561
2013.67		12.74		1558.9		122.40	0.7529

2013.75	12.73	1559.3	122.40	0.7532
2013.83	12.81	1559.7	121.80	0.7570
2013.92	12.93	1560.1	120.60	0.7625
2014.00	13.06	1560.5	119.50	0.7677
2014.08	13.17	1560.9	118.60	0.7720
2014.17	13.24	1561.4	117.90	0.7752
2014.25	13.26	1561.9	117.80	0.7755
2014.33	13.25	1562.4	117.90	0.7745
2014.42	13.26	1562.9	117.90	0.7745
2014.50	13.35	1563.5	117.10	0.7791
2014.58	13.50	1564.1	115.90	0.7864
2014.67	13.59	1564.6	115.10	0.7909
2014.75	13.58	1565.1	115.20	0.7900
2014.83	13.51	1565.6	115.90	0.7863
2014.92	13.43	1566.0	116.60	0.7822
2015.00	13.37	1566.5	117.10	0.7797
2015.08	13.35	1566.9	117.40	0.7785
2015.17	13.31	1567.2	117.80	0.7760
2015.25	13.26	1567.6	118.20	0.7737
2015.33	13.21	1567.9	118.70	0.7712
2015.42	13.14	1568.3	119.40	0.7680
2015.50	13.04	1568.7	120.30	0.7622
2015.58	12.91	1569.0	121.50	0.7553
2015.67	12.86	1569.3	122.00	0.7523
2015.75	12.92	1569.7	121.50	0.7550
2015.83	13.03	1570.1	120.50	0.7596
2015.92	13.13	1570.5	119.60	0.7637
2016.00	13.21	1570.8	118.90	0.7668
2016.08	13.27	1571.1	118.40	0.7691
2016.17	13.33	1571.4	117.90	0.7720
2016.25	13.40	1571.7	117.30	0.7754
2016.33	13.44	1571.9	116.90	0.7771
2016.42	13.48	1572.2	116.60	0.7787
2016.50	13.50	1572.4	116.40	0.7801
2016.58	13.53	1572.6	116.30	0.7816
2016.67	13.55	1572.9	116.10	0.7834
2016.75	13.56	1573.1	116.00	0.7846
2016.83	13.57	1573.4	115.90	0.7858
2016.92	13.59	1573.6	115.80	0.7868
2017.00	13.58	1573.9	115.90	0.7867
2017.08	13.58	1574.2	115.90	0.7866
2017.17	13.60	1574.6	115.70	0.7877
2017.25	13.64	1575.0	115.50	0.7890
2017.33	13.70	1575.4	115.00	0.7915
2017.42	13.76	1575.8	114.50	0.7946
2017.50	13.84	1576.2	113.90	0.7977
2017.58	13.90	1576.6	113.40	0.8005
2017.67	13.91	1577.0	113.30	0.8006
2017.75	13.86	1577.5	113.90	0.7969
2017.83	13.75	1578.0	114.70	0.7914
2017.92	13.67	1578.5	115.50	0.7874
2018.00	13.63	1578.9	115.80	0.7854
2018.08	13.59	1579.3	116.20	0.7833
2018.17	13.53	1579.7	116.70	0.7803

2018.25		13.45		1580.2		117.50	0.7768
2018.33		13.35		1580.7		118.40	0.7723
2018.42		13.24		1581.3		119.40	0.7673
2018.50		13.21		1581.8		119.80	0.7661
2018.58		13.30		1582.3		119.00	0.7714
2018.67		13.41		1582.8		118.00	0.7771
2018.75		13.54		1583.2		116.90	0.7844
2018.83		13.70		1583.5		115.60	0.7923
2018.92		13.81		1583.9		114.70	0.7983
2019.00		13.86		1584.3		114.30	0.8011
2019.08		13.87		1584.7		114.20	0.8027
2019.17		13.88		1585.1		114.20	0.8033
2019.25		13.93		1585.4		113.80	0.8056
2019.33		14.04		1585.7		112.90	0.8108
2019.42		14.16		1586.1		112.00	0.8165
2019.50		14.25		1586.4		111.40	0.8208
2019.58		14.24		1586.6		111.40	0.8203
2019.67		14.20		1587.0		111.80	0.8181
2019.75		14.14		1587.3		112.20	0.8152
2019.83		14.12		1587.8		112.50	0.8132
2019.92		14.12		1588.3		112.50	0.8125
2020.00		14.17		1588.8		112.10	0.8139
2020.08		14.22		1589.2		111.70	0.8158
2020.17		14.28		1589.7		111.30	0.8181
2020.25		14.28		1590.3		111.30	0.8180
2020.33		14.22		1590.8		111.90	0.8145
2020.42		14.14		1591.4		112.60	0.8100
2020.50		14.04		1591.9		113.40	0.8041
2020.58		13.97		1592.5		114.00	0.7996
2020.67		13.93		1593.0		114.40	0.7965
2020.75		13.86		1593.6		115.00	0.7929
2020.83		13.79		1594.2		115.60	0.7894
2020.92		13.73		1594.7		116.20	0.7867
2021.00		13.66		1595.2		116.70	0.7838
2021.08		13.60		1595.7		117.30	0.7808
2021.17		13.54		1596.1		117.90	0.7775
2021.25		13.47		1596.6		118.50	0.7742
2021.33		13.44		1597.0		118.90	0.7720
2021.42		13.45		1597.6		118.80	0.7724
2021.50		13.52		1598.1		118.20	0.7762

Table S2.

year	j-N2O 30S-30N (1e-7/s)							
	fig 1E							
pressure	15 hPa	12 hPa	10 hPa	8.3 hPa	6.8 hPa	5.6 hPa	4.6 hPa	3.8 hPa
2005.08	0.156	0.232	0.333	0.459	0.612	0.787	0.979	1.179
2005.17	0.156	0.233	0.334	0.460	0.613	0.788	0.980	1.180
2005.25	0.156	0.233	0.334	0.461	0.614	0.789	0.980	1.180
2005.33	0.156	0.233	0.334	0.462	0.615	0.789	0.980	1.180
2005.42	0.156	0.233	0.335	0.462	0.615	0.789	0.980	1.179
2005.50	0.156	0.233	0.334	0.461	0.613	0.787	0.977	1.177
2005.58	0.155	0.232	0.333	0.459	0.611	0.785	0.975	1.175
2005.67	0.156	0.233	0.334	0.460	0.612	0.786	0.976	1.176
2005.75	0.156	0.234	0.335	0.461	0.613	0.787	0.977	1.176
2005.83	0.157	0.235	0.336	0.462	0.613	0.787	0.976	1.176
2005.92	0.158	0.235	0.336	0.462	0.613	0.786	0.976	1.175
2006.00	0.158	0.236	0.337	0.462	0.613	0.786	0.975	1.175
2006.08	0.159	0.237	0.337	0.463	0.613	0.786	0.976	1.175
2006.17	0.160	0.237	0.338	0.463	0.614	0.787	0.976	1.176
2006.25	0.160	0.238	0.338	0.463	0.614	0.787	0.977	1.177
2006.33	0.160	0.238	0.338	0.463	0.614	0.787	0.977	1.177
2006.42	0.160	0.238	0.338	0.463	0.614	0.787	0.977	1.177
2006.50	0.161	0.238	0.339	0.464	0.615	0.789	0.979	1.179
2006.58	0.161	0.239	0.339	0.465	0.616	0.791	0.981	1.181
2006.67	0.161	0.238	0.338	0.464	0.615	0.789	0.980	1.180
2006.75	0.160	0.237	0.337	0.463	0.615	0.789	0.980	1.180
2006.83	0.159	0.237	0.337	0.463	0.615	0.789	0.980	1.180
2006.92	0.159	0.236	0.336	0.463	0.615	0.790	0.981	1.181
2007.00	0.158	0.236	0.336	0.463	0.616	0.790	0.982	1.182
2007.08	0.158	0.235	0.336	0.462	0.615	0.790	0.981	1.182
2007.17	0.158	0.235	0.336	0.462	0.615	0.789	0.981	1.181
2007.25	0.158	0.235	0.336	0.462	0.615	0.789	0.980	1.180
2007.33	0.158	0.235	0.337	0.463	0.615	0.788	0.979	1.180
2007.42	0.158	0.236	0.337	0.463	0.615	0.789	0.980	1.180
2007.50	0.158	0.236	0.338	0.464	0.616	0.790	0.981	1.181
2007.58	0.158	0.236	0.338	0.464	0.615	0.789	0.980	1.180
2007.67	0.158	0.236	0.337	0.463	0.615	0.788	0.979	1.179
2007.75	0.158	0.236	0.337	0.463	0.614	0.787	0.978	1.178
2007.83	0.159	0.237	0.338	0.463	0.614	0.787	0.977	1.178
2007.92	0.159	0.238	0.338	0.463	0.614	0.786	0.977	1.178
2008.00	0.160	0.238	0.338	0.463	0.613	0.786	0.977	1.178
2008.08	0.160	0.239	0.339	0.464	0.614	0.787	0.978	1.179
2008.17	0.161	0.239	0.340	0.465	0.615	0.788	0.979	1.180
2008.25	0.161	0.239	0.340	0.465	0.616	0.789	0.980	1.181
2008.33	0.161	0.239	0.339	0.464	0.615	0.789	0.980	1.181
2008.42	0.160	0.238	0.339	0.464	0.615	0.789	0.980	1.182
2008.50	0.160	0.238	0.338	0.463	0.615	0.789	0.980	1.181
2008.58	0.160	0.238	0.338	0.463	0.615	0.789	0.980	1.181
2008.67	0.160	0.238	0.338	0.464	0.616	0.791	0.983	1.184
2008.75	0.160	0.238	0.339	0.465	0.618	0.793	0.985	1.186
2008.83	0.159	0.237	0.339	0.465	0.618	0.793	0.986	1.187
2008.92	0.159	0.237	0.338	0.465	0.618	0.794	0.986	1.187
2009.00	0.158	0.236	0.337	0.465	0.618	0.794	0.986	1.187
2009.08	0.158	0.236	0.337	0.465	0.618	0.794	0.986	1.187
2009.17	0.158	0.236	0.337	0.465	0.618	0.793	0.985	1.186
2009.25	0.158	0.236	0.337	0.465	0.618	0.793	0.985	1.186
2009.33	0.158	0.236	0.338	0.466	0.619	0.793	0.985	1.186
2009.42	0.158	0.236	0.339	0.466	0.619	0.793	0.984	1.185
2009.50	0.158	0.236	0.338	0.465	0.618	0.792	0.983	1.183
2009.58	0.158	0.236	0.338	0.466	0.618	0.791	0.982	1.183
2009.67	0.158	0.236	0.338	0.465	0.617	0.791	0.981	1.182
2009.75	0.157	0.236	0.338	0.465	0.617	0.790	0.981	1.182
2009.83	0.157	0.236	0.339	0.465	0.617	0.790	0.980	1.181
2009.92	0.158	0.236	0.339	0.466	0.617	0.790	0.981	1.182
2010.00	0.158	0.237	0.339	0.466	0.617	0.790	0.980	1.181
2010.08	0.158	0.237	0.339	0.465	0.616	0.788	0.978	1.179
2010.17	0.158	0.236	0.338	0.464	0.615	0.787	0.977	1.178
2010.25	0.158	0.236	0.338	0.464	0.614	0.787	0.977	1.177
2010.33	0.158	0.236	0.338	0.463	0.614	0.786	0.976	1.176
2010.42	0.158	0.236	0.338	0.463	0.613	0.785	0.975	1.175
2010.50	0.158	0.236	0.337	0.462	0.612	0.785	0.975	1.175
2010.58	0.158	0.236	0.336	0.461	0.611	0.784	0.974	1.175
2010.67	0.157	0.235	0.335	0.460	0.610	0.782	0.973	1.174
2010.75	0.157	0.234	0.334	0.459	0.609	0.781	0.971	1.173
2010.83	0.157	0.234	0.333	0.458	0.608	0.781	0.971	1.172

2010.92	0.156	0.233	0.333	0.457	0.608	0.781	0.971	1.172
2011.00	0.156	0.233	0.332	0.457	0.608	0.781	0.971	1.172
2011.08	0.156	0.233	0.333	0.458	0.609	0.782	0.973	1.175
2011.17	0.156	0.233	0.333	0.459	0.610	0.783	0.973	1.175
2011.25	0.156	0.233	0.333	0.459	0.610	0.784	0.974	1.176
2011.33	0.156	0.233	0.333	0.459	0.611	0.784	0.975	1.177
2011.42	0.155	0.232	0.333	0.460	0.611	0.785	0.976	1.178
2011.50	0.156	0.233	0.334	0.461	0.613	0.786	0.977	1.179
2011.58	0.156	0.233	0.335	0.462	0.613	0.787	0.978	1.180
2011.67	0.156	0.234	0.336	0.463	0.615	0.789	0.979	1.181
2011.75	0.156	0.234	0.336	0.464	0.615	0.789	0.979	1.181
2011.83	0.156	0.234	0.337	0.464	0.615	0.788	0.979	1.181
2011.92	0.157	0.235	0.337	0.464	0.615	0.788	0.978	1.180
2012.00	0.157	0.235	0.337	0.464	0.615	0.788	0.978	1.180
2012.08	0.157	0.235	0.337	0.463	0.614	0.786	0.976	1.178
2012.17	0.157	0.235	0.337	0.462	0.613	0.785	0.975	1.177
2012.25	0.157	0.235	0.336	0.461	0.611	0.783	0.973	1.175
2012.33	0.157	0.235	0.336	0.461	0.611	0.783	0.973	1.175
2012.42	0.158	0.236	0.336	0.462	0.612	0.784	0.974	1.175
2012.50	0.158	0.236	0.337	0.462	0.612	0.784	0.974	1.176
2012.58	0.158	0.236	0.337	0.462	0.612	0.784	0.974	1.175
2012.67	0.158	0.236	0.337	0.462	0.612	0.784	0.974	1.175
2012.75	0.159	0.237	0.337	0.461	0.611	0.784	0.974	1.175
2012.83	0.159	0.237	0.337	0.461	0.611	0.783	0.973	1.175
2012.92	0.159	0.237	0.337	0.461	0.610	0.782	0.972	1.174
2013.00	0.159	0.237	0.337	0.461	0.610	0.782	0.972	1.173
2013.08	0.159	0.237	0.337	0.460	0.610	0.782	0.972	1.174
2013.17	0.159	0.237	0.336	0.460	0.609	0.781	0.972	1.174
2013.25	0.159	0.236	0.335	0.459	0.609	0.781	0.972	1.174
2013.33	0.158	0.235	0.334	0.458	0.607	0.780	0.971	1.174
2013.42	0.158	0.234	0.333	0.457	0.606	0.779	0.971	1.173
2013.50	0.158	0.234	0.332	0.456	0.605	0.779	0.970	1.173
2013.58	0.158	0.234	0.332	0.456	0.606	0.780	0.971	1.174
2013.67	0.157	0.233	0.331	0.455	0.605	0.778	0.970	1.173
2013.75	0.156	0.232	0.330	0.454	0.604	0.777	0.969	1.172
2013.83	0.155	0.231	0.329	0.453	0.603	0.777	0.969	1.172
2013.92	0.155	0.230	0.328	0.452	0.603	0.778	0.970	1.173
2014.00	0.154	0.230	0.328	0.452	0.604	0.778	0.971	1.174
2014.08	0.154	0.229	0.328	0.452	0.604	0.778	0.970	1.174
2014.17	0.154	0.229	0.327	0.452	0.604	0.778	0.971	1.174
2014.25	0.154	0.229	0.327	0.452	0.604	0.778	0.970	1.173
2014.33	0.153	0.229	0.328	0.453	0.605	0.779	0.970	1.173
2014.42	0.153	0.228	0.328	0.453	0.604	0.778	0.969	1.172
2014.50	0.153	0.228	0.327	0.452	0.603	0.776	0.967	1.170
2014.58	0.152	0.228	0.327	0.452	0.603	0.776	0.966	1.169
2014.67	0.153	0.228	0.327	0.452	0.603	0.776	0.966	1.169
2014.75	0.153	0.229	0.328	0.454	0.604	0.777	0.967	1.170
2014.83	0.153	0.229	0.329	0.454	0.605	0.777	0.967	1.169
2014.92	0.154	0.230	0.330	0.455	0.605	0.777	0.967	1.169
2015.00	0.154	0.230	0.330	0.455	0.605	0.777	0.967	1.168
2015.08	0.154	0.231	0.330	0.455	0.606	0.778	0.968	1.169
2015.17	0.155	0.231	0.331	0.456	0.606	0.779	0.969	1.170
2015.25	0.155	0.232	0.332	0.457	0.607	0.779	0.969	1.171
2015.33	0.156	0.233	0.333	0.457	0.607	0.779	0.969	1.171
2015.42	0.157	0.233	0.333	0.457	0.607	0.780	0.970	1.172
2015.50	0.157	0.234	0.334	0.458	0.608	0.781	0.972	1.174
2015.58	0.157	0.234	0.334	0.458	0.609	0.782	0.973	1.175
2015.67	0.157	0.234	0.333	0.458	0.609	0.782	0.973	1.175
2015.75	0.156	0.233	0.332	0.456	0.607	0.781	0.972	1.175
2015.83	0.156	0.232	0.331	0.456	0.607	0.781	0.973	1.176
2015.92	0.156	0.232	0.331	0.455	0.606	0.781	0.973	1.176
2016.00	0.156	0.232	0.330	0.454	0.606	0.781	0.973	1.175
2016.08	0.156	0.232	0.330	0.454	0.606	0.780	0.972	1.175
2016.17	0.156	0.231	0.329	0.453	0.604	0.779	0.971	1.174
2016.25	0.156	0.231	0.329	0.453	0.605	0.779	0.971	1.174
2016.33	0.156	0.231	0.329	0.454	0.605	0.780	0.972	1.175
2016.42	0.156	0.231	0.329	0.453	0.605	0.780	0.972	1.175
2016.50	0.155	0.231	0.329	0.453	0.605	0.779	0.972	1.175
2016.58	0.155	0.231	0.329	0.453	0.604	0.779	0.971	1.174
2016.67	0.155	0.231	0.328	0.452	0.604	0.778	0.970	1.173
2016.75	0.156	0.231	0.329	0.453	0.604	0.779	0.971	1.173
2016.83	0.156	0.231	0.329	0.453	0.603	0.778	0.970	1.173
2016.92	0.155	0.231	0.329	0.453	0.604	0.778	0.970	1.173
2017.00	0.155	0.231	0.329	0.453	0.604	0.778	0.971	1.173
2017.08	0.155	0.230	0.329	0.453	0.604	0.778	0.970	1.172

2017.17	0.155	0.230	0.328	0.453	0.604	0.778	0.970	1.173
2017.25	0.154	0.230	0.328	0.453	0.604	0.779	0.971	1.173
2017.33	0.154	0.229	0.328	0.453	0.605	0.779	0.971	1.173
2017.42	0.154	0.230	0.329	0.454	0.605	0.780	0.971	1.173
2017.50	0.154	0.230	0.329	0.455	0.607	0.781	0.972	1.174
2017.58	0.154	0.230	0.330	0.456	0.608	0.782	0.973	1.174
2017.67	0.154	0.231	0.331	0.457	0.608	0.782	0.973	1.174
2017.75	0.155	0.232	0.332	0.458	0.610	0.784	0.974	1.176
2017.83	0.155	0.232	0.333	0.460	0.612	0.785	0.976	1.177
2017.92	0.155	0.233	0.334	0.460	0.612	0.785	0.976	1.177
2018.00	0.156	0.234	0.335	0.461	0.613	0.786	0.976	1.177
2018.08	0.157	0.235	0.336	0.463	0.614	0.787	0.978	1.179
2018.17	0.157	0.236	0.337	0.464	0.615	0.788	0.979	1.180
2018.25	0.158	0.236	0.338	0.464	0.615	0.788	0.978	1.179
2018.33	0.158	0.237	0.339	0.464	0.615	0.787	0.977	1.179
2018.42	0.159	0.237	0.339	0.464	0.615	0.787	0.977	1.178
2018.50	0.158	0.237	0.338	0.463	0.612	0.784	0.974	1.175
2018.58	0.158	0.236	0.337	0.461	0.611	0.783	0.972	1.173
2018.67	0.159	0.236	0.337	0.461	0.611	0.783	0.973	1.173
2018.75	0.159	0.236	0.336	0.460	0.609	0.781	0.971	1.171
2018.83	0.158	0.236	0.335	0.458	0.607	0.779	0.969	1.170
2018.92	0.158	0.235	0.334	0.457	0.606	0.778	0.969	1.170
2019.00	0.158	0.235	0.333	0.456	0.606	0.778	0.968	1.169
2019.08	0.157	0.234	0.332	0.455	0.604	0.776	0.966	1.167
2019.17	0.157	0.233	0.331	0.454	0.604	0.776	0.966	1.167
2019.25	0.156	0.231	0.329	0.453	0.602	0.775	0.966	1.166
2019.33	0.155	0.231	0.329	0.452	0.603	0.776	0.967	1.167
2019.42	0.155	0.230	0.328	0.452	0.603	0.777	0.967	1.168
2019.50	0.155	0.230	0.328	0.453	0.604	0.778	0.969	1.170
2019.58	0.155	0.230	0.328	0.453	0.605	0.779	0.970	1.171
2019.67	0.154	0.230	0.328	0.453	0.605	0.779	0.970	1.171
2019.75	0.154	0.230	0.329	0.454	0.606	0.780	0.971	1.172
2019.83	0.154	0.229	0.329	0.455	0.607	0.781	0.972	1.173
2019.92	0.154	0.230	0.329	0.455	0.608	0.782	0.972	1.173
2020.00	0.154	0.230	0.329	0.455	0.608	0.782	0.972	1.173
2020.08	0.154	0.230	0.330	0.456	0.608	0.781	0.972	1.172
2020.17	0.154	0.230	0.330	0.456	0.607	0.781	0.971	1.172
2020.25	0.155	0.231	0.332	0.457	0.609	0.782	0.972	1.173
2020.33	0.155	0.232	0.332	0.458	0.608	0.781	0.971	1.172
2020.42	0.156	0.233	0.333	0.458	0.608	0.781	0.971	1.171
2020.50	0.156	0.233	0.333	0.458	0.609	0.781	0.971	1.171
2020.58	0.156	0.233	0.333	0.458	0.608	0.780	0.970	1.170
2020.67	0.156	0.233	0.333	0.459	0.609	0.781	0.971	1.172
2020.75	0.156	0.233	0.334	0.459	0.610	0.782	0.972	1.172
2020.83	0.156	0.233	0.334	0.459	0.610	0.782	0.972	1.172
2020.92	0.156	0.233	0.334	0.459	0.610	0.782	0.972	1.172
2021.00	0.156	0.233	0.334	0.459	0.610	0.783	0.972	1.173
2021.08	0.156	0.234	0.334	0.460	0.611	0.783	0.973	1.174
2021.17	0.157	0.234	0.335	0.461	0.612	0.784	0.974	1.174
2021.25	0.157	0.234	0.335	0.461	0.612	0.784	0.974	1.174
2021.33	0.157	0.234	0.335	0.461	0.612	0.784	0.974	1.174
2021.42	0.157	0.234	0.335	0.461	0.612	0.783	0.973	1.173
2021.50	0.157	0.235	0.335	0.461	0.611	0.783	0.973	1.173

Table S3.

year	f-N2O 30S-30N (ppb)						
	fig 1F						
	32 hPa	22 hPa	15 hPa	10 hPa	6.8 hPa	4.6 hPa	3.2 hPa
2005.08	209.19	203.02	196.73	163.04	119.83	78.55	47.37
2005.17	209.30	202.90	197.61	163.83	118.97	77.17	47.20
2005.25	209.53	202.84	198.29	164.44	118.00	75.29	46.34
2005.33	210.19	203.06	198.64	164.67	116.99	73.36	45.12
2005.42	210.93	203.63	199.30	164.96	116.25	71.88	43.89
2005.50	211.78	204.24	199.80	165.25	116.48	72.45	44.72
2005.58	212.87	204.81	199.93	165.33	117.16	74.19	47.06
2005.67	214.05	205.29	199.55	164.47	116.87	75.31	49.26
2005.75	215.03	205.43	198.20	162.12	115.07	75.17	50.48
2005.83	216.01	205.67	196.80	159.30	112.59	74.39	51.05
2005.92	216.89	205.97	195.42	156.28	109.81	73.27	51.23
2006.00	217.68	206.34	194.12	153.47	107.20	71.93	50.87
2006.08	218.10	206.40	192.79	151.46	105.56	70.59	49.67
2006.17	218.21	206.21	191.61	150.28	104.91	69.46	47.79
2006.25	218.25	205.85	190.38	149.52	105.24	69.35	46.34
2006.33	218.06	205.31	189.33	148.94	105.75	69.85	45.80
2006.42	217.48	204.58	188.34	148.47	106.29	70.64	45.99
2006.50	217.04	204.33	188.12	148.37	106.18	70.00	44.88
2006.58	216.46	204.26	188.69	149.11	106.30	68.83	42.75
2006.67	215.84	204.38	189.93	150.80	107.29	68.30	40.91
2006.75	215.23	204.73	192.06	153.87	109.56	68.86	40.05
2006.83	214.40	204.79	194.07	157.19	112.22	69.78	39.79
2006.92	213.76	204.92	195.97	160.50	115.00	70.90	39.76
2007.00	213.37	205.02	197.43	163.09	117.37	72.20	40.15
2007.08	213.39	205.48	198.78	164.54	118.60	73.49	41.36
2007.17	213.76	206.55	200.19	164.97	118.49	74.40	43.25
2007.25	214.34	207.90	201.78	164.89	117.02	73.99	44.68
2007.33	214.92	209.30	203.48	165.09	115.59	73.00	45.28
2007.42	215.61	210.61	204.94	165.38	114.33	71.49	44.75
2007.50	216.19	211.27	205.28	165.22	113.88	71.07	44.42
2007.58	216.71	211.46	204.55	164.18	113.64	72.03	45.80
2007.67	217.35	211.55	203.51	162.86	113.35	73.32	48.23
2007.75	218.09	211.54	202.09	160.78	112.17	73.92	50.30
2007.83	219.00	211.66	200.54	158.17	110.35	73.99	51.69
2007.92	219.86	211.79	198.97	155.31	108.17	73.62	52.47
2008.00	220.42	212.09	197.81	152.87	106.21	73.09	52.80
2008.08	220.63	212.09	196.70	151.25	105.03	72.37	52.13
2008.17	220.48	211.43	195.63	150.63	104.75	71.54	50.52
2008.25	220.00	210.44	194.71	150.88	105.62	71.40	49.01
2008.33	219.51	209.39	193.77	151.41	107.39	72.54	48.53
2008.42	219.00	208.40	192.98	151.90	109.24	74.28	49.04
2008.50	218.32	207.76	192.96	152.73	110.46	75.19	49.46
2008.58	217.40	207.31	193.91	154.33	111.26	74.61	48.24
2008.67	216.36	206.75	195.29	156.63	112.43	73.73	45.85
2008.75	214.87	206.00	196.61	158.97	113.58	72.59	43.09
2008.83	213.28	205.11	197.87	161.60	115.25	72.01	41.08
2008.92	211.93	204.18	199.01	164.45	117.49	72.26	40.00
2009.00	210.94	203.34	200.10	167.28	119.85	72.84	39.43
2009.08	210.39	202.98	201.03	169.17	121.25	73.21	39.36
2009.17	210.16	203.16	201.70	169.74	121.37	73.25	39.75
2009.25	210.19	203.79	202.42	169.47	120.35	72.76	40.26
2009.33	210.23	204.62	203.38	169.00	118.42	71.10	39.89
2009.42	210.25	205.46	204.57	169.04	117.04	69.57	39.16
2009.50	210.48	206.08	205.64	169.41	116.59	69.39	39.63
2009.58	211.01	206.81	206.29	169.24	115.86	69.34	40.64
2009.67	211.78	207.73	206.60	168.68	114.80	68.98	41.64
2009.75	212.88	208.86	207.04	168.24	113.68	68.30	42.15
2009.83	214.12	210.12	207.68	168.05	112.68	67.42	42.09
2009.92	215.29	211.39	208.16	167.63	111.72	66.46	41.55
2010.00	216.40	212.64	208.20	166.57	110.51	65.63	41.15
2010.08	217.26	213.51	208.03	165.44	109.67	65.74	41.72
2010.17	218.01	214.20	208.02	164.84	109.64	66.91	43.41
2010.25	218.52	214.39	207.57	164.04	109.44	67.72	44.76
2010.33	218.81	214.16	206.70	163.07	109.13	68.37	46.05
2010.42	218.84	213.65	205.50	161.89	108.62	68.78	47.08
2010.50	218.76	213.36	204.55	161.32	108.96	69.34	47.23
2010.58	218.53	212.98	203.98	161.73	110.69	71.38	48.33
2010.67	217.62	212.11	203.53	162.46	112.67	73.64	50.01
2010.75	216.55	211.02	203.17	163.58	115.25	76.47	52.07

2010.83	215.28	209.69	202.88	164.72	117.60	78.77	53.33
2010.92	213.87	208.29	202.86	166.02	119.58	80.76	54.79
2011.00	212.45	206.76	202.83	167.27	121.11	82.16	55.92
2011.08	211.34	205.49	202.82	168.31	122.00	82.49	56.02
2011.17	210.40	204.50	202.74	169.09	122.40	81.96	55.07
2011.25	209.62	203.97	202.94	169.96	122.93	81.50	54.08
2011.33	208.84	203.60	203.33	170.77	123.34	81.07	53.01
2011.42	208.21	203.42	203.99	171.76	123.82	80.77	52.15
2011.50	207.62	203.14	204.51	171.99	122.96	79.44	51.21
2011.58	207.37	202.96	204.94	171.93	121.58	77.45	49.78
2011.67	207.66	203.30	205.22	171.19	119.58	74.92	47.45
2011.75	208.04	203.98	205.52	170.35	117.55	72.42	45.38
2011.83	208.68	205.06	205.66	169.31	115.92	70.91	44.46
2011.92	209.36	206.25	205.50	167.81	114.27	69.58	43.41
2012.00	209.93	207.37	205.28	166.29	112.81	68.63	42.69
2012.08	210.34	208.31	205.17	165.17	111.87	68.31	42.71
2012.17	210.83	209.02	205.12	164.37	111.30	68.41	43.19
2012.25	211.43	209.47	204.88	163.58	110.88	68.71	43.82
2012.33	212.29	209.99	204.46	162.61	110.34	68.90	44.37
2012.42	213.25	210.57	203.96	161.18	108.93	68.20	44.40
2012.50	214.21	210.88	203.02	159.65	107.53	67.15	43.75
2012.58	215.17	211.05	201.71	158.41	107.61	67.74	43.79
2012.67	216.04	210.78	199.67	156.72	108.02	69.32	45.00
2012.75	217.03	210.42	197.30	154.31	107.81	70.97	46.66
2012.83	217.82	210.05	195.24	151.70	106.70	71.91	48.22
2012.92	218.78	209.95	193.86	149.65	105.31	72.12	49.44
2013.00	219.58	209.91	193.02	148.29	104.10	71.85	49.98
2013.08	220.18	209.84	192.43	147.77	103.99	71.89	49.79
2013.17	220.45	209.59	191.84	147.84	105.03	72.96	49.93
2013.25	220.50	209.25	191.40	148.43	107.02	75.17	51.00
2013.33	220.23	208.63	190.87	149.20	109.44	77.97	52.69
2013.42	219.90	207.89	190.24	150.08	112.19	81.07	54.61
2013.50	219.27	207.20	189.76	150.61	113.99	83.22	56.15
2013.58	218.27	206.74	190.01	150.92	113.64	82.38	55.43
2013.67	217.09	206.48	191.17	152.24	113.80	81.52	54.29
2013.75	215.91	206.13	192.68	154.48	115.16	81.66	53.74
2013.83	214.82	205.77	194.07	157.14	117.50	82.64	53.61
2013.92	213.56	205.13	194.94	159.64	120.26	84.06	53.67
2014.00	212.47	204.62	195.65	161.83	122.83	85.53	53.93
2014.08	211.55	204.45	196.57	163.53	124.22	86.19	54.37
2014.17	210.71	204.41	197.68	164.99	124.68	85.74	54.22
2014.25	210.12	204.63	198.83	166.22	124.57	84.38	53.15
2014.33	209.70	205.23	200.31	167.54	124.37	82.79	51.65
2014.42	209.28	206.01	202.02	168.81	124.20	81.55	50.43
2014.50	209.11	206.95	203.92	170.29	124.62	81.55	50.76
2014.58	209.33	207.80	205.20	171.04	124.84	82.19	52.25
2014.67	209.60	208.81	206.21	170.94	123.98	81.99	53.32
2014.75	209.87	209.90	206.91	170.02	121.83	80.28	52.97
2014.83	210.42	211.08	207.65	169.07	119.29	77.89	52.03
2014.92	211.14	212.17	208.35	168.24	116.93	75.45	50.68
2015.00	211.90	212.69	208.30	167.52	115.78	74.17	49.66
2015.08	212.62	212.63	207.22	166.55	115.77	74.32	49.17
2015.17	213.22	212.31	205.42	164.76	115.61	75.07	49.20
2015.25	213.43	211.79	203.36	162.35	114.93	76.07	50.04
2015.33	213.44	211.24	201.28	159.66	113.72	76.84	51.31
2015.42	213.19	210.60	199.42	157.39	112.47	77.07	52.19
2015.50	212.77	210.01	198.27	156.33	111.82	76.46	51.34
2015.58	212.00	209.17	197.60	156.52	112.29	75.92	49.52
2015.67	211.49	208.13	197.04	157.76	114.55	77.00	48.51
2015.75	211.43	207.01	196.42	159.62	118.28	79.99	49.27
2015.83	211.30	205.66	195.19	160.92	121.94	83.19	50.37
2015.92	211.11	204.38	193.75	161.57	124.88	86.23	51.90
2016.00	210.77	203.47	192.81	161.80	126.56	88.26	53.34
2016.08	210.40	202.95	192.59	161.81	126.84	88.98	54.37
2016.17	210.05	202.66	192.96	162.15	126.87	89.31	55.24
2016.25	209.80	202.26	193.47	162.92	127.18	89.44	55.69
2016.33	209.77	201.69	193.67	163.64	127.58	89.36	55.52
2016.42	210.09	201.16	193.63	164.18	128.20	89.73	55.67
2016.50	210.45	200.80	193.25	164.05	128.59	90.44	56.24
2016.58	210.66	200.70	192.88	163.65	128.71	91.15	57.11
2016.67	210.76	200.87	192.77	163.24	128.51	91.51	57.94
2016.75	210.36	201.18	192.96	162.75	127.82	91.26	58.39
2016.83	209.51	201.38	193.68	162.73	127.31	91.35	59.17
2016.92	208.51	201.51	194.71	163.20	127.16	91.48	59.68

2017.00	207.56	201.72	195.87	164.04	127.28	91.47	59.80
2017.08	206.87	202.08	197.16	165.46	128.02	91.52	59.64
2017.17	206.44	202.57	198.47	167.18	129.27	91.70	59.26
2017.25	206.12	203.32	199.88	169.03	130.66	91.79	58.53
2017.33	205.74	204.36	201.68	171.02	131.88	91.67	57.74
2017.42	205.23	205.54	203.68	172.74	132.55	91.29	56.99
2017.50	204.74	206.61	205.56	173.83	132.35	90.60	56.62
2017.58	204.33	207.62	207.24	174.23	131.14	89.44	56.63
2017.67	204.17	208.35	208.53	174.24	129.31	87.50	56.10
2017.75	204.16	208.95	209.63	174.10	127.04	84.57	54.30
2017.83	204.40	209.84	210.69	173.77	124.50	81.02	51.65
2017.92	204.90	210.91	211.63	173.49	122.47	78.09	49.31
2018.00	205.51	212.01	212.29	172.72	120.46	75.91	47.77
2018.08	205.85	212.88	212.51	171.16	117.99	74.08	46.84
2018.17	206.22	213.74	212.56	169.16	115.09	72.13	46.13
2018.25	206.79	214.65	212.70	167.19	112.07	70.21	45.78
2018.33	207.40	215.10	212.46	165.31	109.37	68.49	45.55
2018.42	207.96	214.96	211.84	163.95	107.45	66.94	44.97
2018.50	208.72	214.55	211.14	163.77	107.78	67.39	45.26
2018.58	209.66	214.10	210.12	163.97	109.96	70.17	47.12
2018.67	210.14	213.33	208.32	163.46	112.01	73.08	49.09
2018.75	210.34	212.39	206.26	162.55	113.85	76.41	51.84
2018.83	210.53	211.33	204.32	161.95	116.21	80.49	55.16
2018.92	210.63	210.21	202.60	161.65	118.37	83.99	58.10
2019.00	210.79	209.32	201.42	161.89	120.29	86.45	60.10
2019.08	210.89	208.68	200.97	162.81	122.16	88.08	61.13
2019.17	210.75	208.23	201.22	164.38	124.04	89.06	61.23
2019.25	210.68	207.84	201.90	166.79	126.76	90.38	61.13
2019.33	210.56	207.68	202.79	169.52	129.84	91.90	61.07
2019.42	210.56	207.92	203.56	171.46	132.19	93.41	61.51
2019.50	210.22	208.38	204.23	172.30	132.94	93.80	61.78
2019.58	209.48	208.83	205.22	173.01	132.47	92.28	60.46
2019.67	208.58	209.61	207.03	174.27	132.21	90.81	58.71
2019.75	207.74	210.34	209.14	176.05	132.37	89.33	56.55
2019.83	207.15	210.94	211.24	178.01	132.32	87.43	54.34
2019.92	206.84	211.45	213.01	179.48	132.04	85.85	52.68
2020.00	206.55	211.77	214.30	180.28	131.53	84.81	51.91
2020.08	206.24	211.82	214.80	180.21	130.82	84.34	51.93
2020.17	205.92	211.69	214.79	179.62	130.11	84.56	52.83
2020.25	205.38	211.25	214.16	178.24	128.58	84.15	53.57
2020.33	204.91	210.72	213.30	176.62	126.77	83.33	53.90
2020.42	204.24	210.05	212.51	175.53	125.56	82.47	53.64
2020.50	203.76	209.58	212.07	174.96	124.71	81.45	52.62
2020.58	203.65	209.33	212.21	175.31	124.88	81.30	51.93
2020.67	203.52	208.97	212.44	175.87	125.02	80.90	51.23
2020.75	203.39	208.67	212.42	176.00	124.79	80.17	50.35
2020.83	202.93	208.38	212.21	175.81	124.62	79.78	49.82
2020.92	202.28	208.21	212.20	175.77	124.58	79.68	49.61
2021.00	201.42	207.95	212.31	175.94	124.64	79.44	49.12
2021.08	200.92	208.05	212.65	176.32	124.63	78.78	48.26
2021.17	200.61	208.18	212.88	176.50	124.20	77.57	47.08
2021.25	200.34	208.62	213.24	176.34	123.39	76.18	45.78
2021.33	200.06	209.29	213.79	176.13	122.69	75.16	44.69
2021.42	200.06	210.14	214.50	176.11	122.41	74.84	44.18
2021.50	200.08	210.68	214.73	175.64	122.18	75.40	45.07

Table S4.

year	f-N2O 30N-58N (ppb)						
no figure							
pressure	32 hPa	22 hPa	15 hPa	10 hPa	6.8 hPa	4.6 hPa	3.2 hPa
2005.08	140.94	131.71	112.79	76.69	48.41	31.91	21.18
2005.17	139.81	131.22	114.28	79.30	50.45	32.92	21.61
2005.25	138.73	130.53	115.31	81.54	52.64	34.44	22.54
2005.33	138.25	130.05	115.93	83.31	54.69	35.96	23.34
2005.42	137.58	129.74	116.79	84.94	55.74	35.96	22.92
2005.50	136.92	129.71	116.92	84.01	53.76	34.26	22.33
2005.58	136.98	129.55	115.38	81.18	51.27	33.35	22.67
2005.67	138.18	129.58	113.49	78.25	48.99	32.61	23.06
2005.75	140.82	130.87	112.42	75.52	46.50	31.38	22.85
2005.83	143.67	132.51	111.72	73.26	44.46	30.32	22.46
2005.92	146.07	133.86	111.04	71.13	42.51	29.33	22.16
2006.00	148.42	135.52	110.92	69.52	40.74	28.33	21.85
2006.08	150.55	137.31	111.16	68.24	39.01	27.14	21.33
2006.17	152.56	139.31	111.85	67.42	37.46	25.78	20.51
2006.25	154.56	141.32	112.68	66.96	36.18	24.36	19.40
2006.33	156.25	143.21	113.66	66.58	34.86	22.93	18.40
2006.42	158.26	145.21	114.42	65.96	34.01	22.69	18.55
2006.50	159.62	145.78	114.77	67.27	36.14	24.36	19.09
2006.58	159.73	145.99	116.50	70.19	38.46	24.99	18.49
2006.67	158.30	145.87	118.56	73.13	40.57	25.60	18.11
2006.75	156.11	144.99	119.74	75.57	42.70	26.65	18.29
2006.83	153.77	143.92	120.99	78.08	44.63	27.53	18.62
2006.92	151.26	142.62	121.91	80.32	46.51	28.39	18.86
2007.00	149.05	141.39	122.44	82.04	48.17	29.24	19.08
2007.08	146.90	139.92	122.64	83.51	49.80	30.30	19.54
2007.17	144.93	138.25	122.27	84.38	51.17	31.48	20.27
2007.25	143.04	136.61	121.77	84.89	52.25	32.65	21.18
2007.33	141.47	135.21	121.23	85.29	53.24	33.70	22.01
2007.42	139.72	133.93	120.85	85.66	53.75	33.85	21.89
2007.50	138.91	134.23	121.50	85.49	52.69	32.79	21.42
2007.58	139.85	135.28	120.76	83.53	51.23	32.55	21.89
2007.67	141.34	135.21	118.46	80.82	49.70	32.44	22.56
2007.75	142.50	134.54	115.89	78.04	48.07	32.10	22.97
2007.83	143.55	134.12	113.35	75.08	46.36	31.66	22.99
2007.92	145.20	134.39	111.31	72.32	44.62	31.15	22.97
2008.00	146.57	134.68	109.73	69.93	42.90	30.51	22.91
2008.08	147.93	135.29	108.52	67.66	41.02	29.56	22.58
2008.17	149.25	136.12	107.82	65.80	39.15	28.33	21.93
2008.25	150.50	136.97	107.41	64.38	37.45	26.89	20.90
2008.33	151.55	137.80	107.36	63.32	35.87	25.41	19.84
2008.42	152.41	138.28	106.98	62.30	35.04	25.19	19.98
2008.50	152.44	137.38	105.63	61.74	35.69	26.19	20.48
2008.58	150.73	135.32	105.63	63.91	38.00	27.15	20.31
2008.67	147.93	134.17	107.78	67.50	40.50	27.67	19.55
2008.75	146.18	134.52	111.33	71.59	42.54	27.41	18.31
2008.83	144.76	135.05	114.93	75.63	44.53	27.42	17.72
2008.92	143.27	135.01	117.83	79.36	46.65	27.81	17.52
2009.00	141.82	134.42	119.91	82.71	48.94	28.51	17.48
2009.08	140.47	133.44	121.07	85.41	51.28	29.62	17.74

2009.17	139.20	132.29	121.54	87.43	53.48	31.02	18.33
2009.25	138.11	131.17	121.57	88.78	55.37	32.64	19.41
2009.33	137.22	130.12	121.22	89.62	57.03	34.35	20.64
2009.42	136.28	128.80	120.60	90.07	57.75	34.59	20.47
2009.50	135.10	128.26	121.34	90.59	56.69	32.84	19.37
2009.58	134.75	128.82	121.78	89.37	54.44	31.32	19.13
2009.67	135.41	130.12	122.28	88.57	53.05	30.60	19.47
2009.75	136.47	131.24	122.52	88.19	52.52	30.47	19.89
2009.83	137.53	132.09	122.54	87.96	52.30	30.39	20.01
2009.92	138.54	133.15	122.82	87.98	52.34	30.36	19.92
2010.00	139.34	134.46	123.38	88.13	52.49	30.51	19.97
2010.08	140.00	135.82	124.28	88.51	52.74	30.76	20.16
2010.17	140.57	137.26	125.46	89.12	53.09	31.11	20.51
2010.25	141.06	138.74	126.86	89.92	53.46	31.34	20.71
2010.33	141.56	140.17	128.31	90.58	53.32	30.93	20.46
2010.42	142.51	141.89	129.48	90.42	52.54	30.56	20.75
2010.50	143.33	141.56	127.30	88.49	52.67	32.06	22.14
2010.58	142.56	138.89	124.44	87.66	53.79	33.58	23.03
2010.67	140.85	135.37	121.40	86.61	54.66	35.08	24.18
2010.75	138.00	131.32	117.93	85.04	55.10	36.50	25.62
2010.83	134.17	127.18	115.31	84.31	55.72	37.44	26.29
2010.92	130.75	123.28	112.76	83.40	55.97	38.10	26.85
2011.00	127.93	119.62	109.97	81.95	55.58	38.22	27.10
2011.08	125.58	116.14	107.04	80.25	54.88	37.98	27.01
2011.17	123.48	112.81	104.01	78.33	53.94	37.55	26.74
2011.25	121.31	109.55	100.99	76.32	52.87	37.02	26.47
2011.33	119.31	106.49	97.94	74.40	52.28	37.11	26.57
2011.42	117.54	103.99	95.92	74.17	53.43	38.28	26.94
2011.50	116.27	103.14	96.83	76.08	54.67	38.37	26.45
2011.58	115.87	104.93	99.55	77.46	54.27	37.27	25.52
2011.67	116.69	108.02	102.33	78.01	53.08	35.60	23.98
2011.75	118.00	110.77	104.39	78.43	52.26	34.23	22.52
2011.83	119.91	113.39	105.60	77.98	51.17	33.28	21.80
2011.92	121.46	115.75	106.77	77.61	50.22	32.48	21.23
2012.00	122.46	117.71	108.09	77.71	49.69	31.96	20.89
2012.08	123.13	119.51	109.60	78.18	49.48	31.68	20.76
2012.17	123.72	121.05	111.10	78.91	49.53	31.47	20.60
2012.25	124.33	122.28	112.47	79.84	49.92	31.50	20.46
2012.33	124.64	123.39	114.06	81.14	50.45	31.42	20.28
2012.42	124.94	124.92	116.32	82.58	50.44	30.64	19.62
2012.50	126.02	127.04	117.76	82.39	49.35	29.70	19.20
2012.58	128.26	127.90	116.26	80.20	48.17	29.52	19.32
2012.67	130.89	127.52	113.23	77.57	47.25	29.48	19.34
2012.75	133.27	126.61	110.08	75.18	46.88	30.43	20.51
2012.83	135.88	125.93	107.23	73.08	46.49	31.04	21.36
2012.92	138.34	125.39	104.42	70.65	45.52	31.03	21.70
2013.00	140.93	125.57	102.18	68.32	44.44	30.89	21.85
2013.08	143.52	126.29	100.62	66.21	43.14	30.44	21.76
2013.17	145.84	127.41	99.77	64.48	41.75	29.73	21.45
2013.25	148.09	128.75	99.46	63.18	40.37	28.72	20.83
2013.33	150.04	129.92	99.29	61.99	38.97	27.73	20.35
2013.42	151.30	130.27	98.42	60.53	37.92	27.60	20.84
2013.50	151.92	130.44	98.50	61.12	39.17	28.97	21.80
2013.58	151.38	130.41	100.14	64.09	41.95	30.52	22.25

2013.67	149.48	130.51	103.26	68.06	44.58	31.61	22.57
2013.75	147.38	131.42	107.06	71.50	45.83	31.28	21.83
2013.83	144.96	131.88	110.03	74.05	46.80	31.37	21.68
2013.92	142.37	131.97	112.71	76.75	48.17	31.77	21.71
2014.00	139.87	131.66	114.93	79.35	49.63	32.17	21.68
2014.08	137.38	130.94	116.52	81.69	51.25	32.80	21.80
2014.17	135.06	129.92	117.45	83.54	52.77	33.60	22.13
2014.25	132.90	128.87	117.91	84.73	53.92	34.45	22.70
2014.33	131.15	127.98	117.97	85.45	54.94	35.36	23.26
2014.42	129.67	126.94	117.64	85.98	55.78	35.61	22.79
2014.50	128.10	125.69	117.17	85.67	54.69	33.98	21.56
2014.58	126.95	125.78	117.71	84.84	52.65	32.45	21.39
2014.67	127.29	126.88	117.72	82.94	50.37	31.56	21.97
2014.75	128.14	128.13	118.07	82.12	49.27	31.07	22.18
2014.83	128.37	129.03	118.96	82.44	48.78	30.26	21.67
2014.92	128.78	129.93	119.84	82.94	48.68	29.82	21.32
2015.00	129.25	130.88	120.70	83.42	48.74	29.68	21.21
2015.08	129.71	131.64	121.31	83.79	48.86	29.62	21.09
2015.17	130.10	132.19	121.68	84.03	49.03	29.61	20.95
2015.25	130.49	132.84	122.14	84.42	49.35	29.71	20.86
2015.33	130.90	133.59	122.88	84.97	49.55	29.59	20.65
2015.42	131.59	134.66	123.80	85.13	49.23	29.61	21.19
2015.50	132.11	134.96	123.38	84.75	49.99	31.24	22.64
2015.58	132.18	134.12	122.38	85.27	51.85	32.76	22.89
2015.67	130.74	132.11	121.82	87.05	54.34	33.96	22.44
2015.75	128.13	129.38	120.64	87.70	55.80	34.87	22.31
2015.83	126.52	127.97	120.08	88.24	57.07	35.91	22.55
2015.92	125.21	126.79	119.67	88.67	57.96	36.60	22.73
2016.00	124.03	125.45	119.07	88.88	58.61	37.20	22.99
2016.08	122.92	124.18	118.49	88.95	59.04	37.71	23.37
2016.17	122.05	123.18	118.08	89.06	59.48	38.32	23.94
2016.25	121.27	122.25	117.60	88.93	59.62	38.72	24.49
2016.33	120.58	121.46	117.15	88.75	59.64	38.97	24.83
2016.42	119.87	120.24	115.96	87.90	59.29	38.94	24.75
2016.50	118.71	119.22	115.62	88.10	59.60	38.92	24.37
2016.58	117.63	118.53	115.44	87.96	59.38	38.79	24.45
2016.67	116.86	117.89	114.98	87.71	59.22	38.67	24.50
2016.75	116.67	117.50	114.40	87.32	59.02	38.56	24.57
2016.83	116.42	116.76	113.38	86.39	58.54	38.52	24.80
2016.92	116.02	116.01	112.48	85.66	58.22	38.58	25.01
2017.00	115.46	115.33	111.61	84.88	57.80	38.48	25.05
2017.08	114.80	114.66	110.86	84.20	57.34	38.32	25.07
2017.17	114.10	113.99	110.21	83.61	56.85	37.99	24.91
2017.25	113.19	113.16	109.56	83.17	56.50	37.71	24.73
2017.33	112.00	112.13	108.76	82.74	56.48	37.94	24.97
2017.42	110.91	111.49	108.74	83.56	57.72	38.79	25.16
2017.50	110.29	111.35	109.40	84.56	58.22	38.64	24.79
2017.58	110.28	112.76	111.50	85.76	57.91	37.78	24.35
2017.67	111.16	115.07	113.60	86.06	56.83	36.90	24.37
2017.75	112.85	118.72	117.11	87.49	56.35	36.01	24.01
2017.83	114.57	122.04	120.30	89.05	56.15	35.13	23.34
2017.92	116.33	125.26	123.16	90.43	56.13	34.57	22.90
2018.00	117.88	128.20	125.97	92.04	56.48	34.30	22.61
2018.08	119.16	130.90	128.66	93.75	57.13	34.28	22.43

2018.17	120.35	133.48	131.33	95.62	58.17	34.71	22.50
2018.25	121.76	135.96	133.87	97.51	59.36	35.35	22.74
2018.33	123.46	138.52	136.57	99.58	60.50	35.63	22.57
2018.42	125.55	141.47	139.08	100.48	59.89	34.64	22.08
2018.50	127.94	143.32	138.97	98.48	57.55	33.37	21.95
2018.58	129.77	141.78	134.74	94.13	55.34	33.14	22.28
2018.67	130.84	138.26	129.41	91.08	55.54	34.47	23.07
2018.75	130.64	133.30	123.82	88.90	56.50	36.21	24.42
2018.83	129.90	127.93	118.08	86.42	57.19	38.12	26.23
2018.92	128.98	122.66	112.12	83.33	57.15	39.50	27.70
2019.00	128.64	118.45	106.63	79.81	56.38	40.30	28.85
2019.08	128.93	115.17	101.71	76.18	55.09	40.52	29.50
2019.17	129.37	112.51	97.16	72.21	53.00	39.97	29.63
2019.25	129.66	110.37	93.52	68.75	50.73	38.91	29.32
2019.33	129.52	108.34	90.26	65.53	48.52	37.91	29.14
2019.42	128.57	106.25	87.94	64.16	48.58	38.74	29.80
2019.50	126.72	104.70	87.82	65.54	50.16	39.41	29.78
2019.58	125.29	105.70	90.98	69.00	51.98	39.65	29.77
2019.67	124.26	108.51	95.22	71.23	51.78	38.75	29.34
2019.75	123.96	111.66	98.68	72.59	51.96	38.75	29.02
2019.83	123.67	114.77	102.55	75.04	53.22	39.38	28.90
2019.92	123.34	117.80	106.51	77.64	54.47	39.82	28.74
2020.00	122.62	120.25	110.16	80.23	55.75	40.29	28.66
2020.08	121.51	122.16	113.54	82.81	57.03	40.79	28.77
2020.17	120.23	123.58	116.66	85.51	58.52	41.45	29.04
2020.25	119.01	124.61	119.05	87.74	60.01	42.29	29.48
2020.33	118.03	125.41	120.89	89.50	61.23	43.00	29.78
2020.42	117.77	126.34	122.07	89.99	60.94	42.41	29.38
2020.50	118.26	127.40	122.63	89.58	60.30	42.27	29.51
2020.58	117.82	126.46	121.26	88.67	60.32	42.54	29.28
2020.67	117.47	126.14	121.11	88.82	60.54	42.38	28.56
2020.75	117.03	126.13	121.80	89.41	60.13	41.16	27.34
2020.83	117.00	126.72	122.50	89.23	58.87	39.54	26.23
2020.92	116.94	127.21	123.26	89.20	57.87	38.40	25.58
2021.00	116.51	127.22	123.82	89.53	57.40	37.62	25.10
2021.08	116.19	127.01	124.01	89.77	57.20	37.08	24.66
2021.17	116.01	126.72	123.97	90.01	57.31	36.87	24.33
2021.25	115.70	126.29	123.75	90.13	57.52	36.96	24.29
2021.33	115.50	125.75	123.29	90.15	57.89	37.18	24.19
2021.42	114.76	124.68	123.01	90.89	58.52	37.06	23.71
2021.50	114.02	124.22	123.38	91.10	57.82	36.04	23.12

Table S5.

year	f-N2O 30S-58S (ppb)						
no figure							
pressure	32 hPa	22 hPa	15 hPa	10 hPa	6.8 hPa	4.6 hPa	3.2 hPa
2005.08	131.23	109.32	99.84	79.50	56.59	38.02	24.24
2005.17	129.96	109.84	101.95	80.84	55.99	36.77	23.77
2005.25	129.54	111.23	104.00	81.56	54.90	35.20	23.02
2005.33	129.41	112.64	105.96	82.80	54.58	33.81	21.79
2005.42	129.09	113.96	108.14	84.52	54.98	33.08	20.85
2005.50	128.90	115.31	110.41	86.41	55.83	33.03	20.44
2005.58	128.54	116.36	112.57	88.47	57.08	33.53	20.50
2005.67	128.16	117.30	114.48	90.38	58.38	34.24	20.84
2005.75	127.93	118.21	116.24	92.22	59.71	35.04	21.31
2005.83	127.90	119.54	118.46	94.40	61.24	35.87	21.66
2005.92	128.73	121.43	120.88	95.99	61.36	35.14	20.99
2006.00	130.54	123.72	122.53	95.78	59.65	33.37	19.88
2006.08	133.34	125.64	122.50	94.26	58.22	32.65	19.50
2006.17	135.88	125.92	120.80	92.88	58.30	33.11	19.34
2006.25	137.44	125.12	118.75	91.83	58.71	33.62	19.02
2006.33	138.61	123.80	116.29	90.37	58.95	34.55	19.41
2006.42	139.72	122.87	114.07	88.63	58.86	35.57	20.29
2006.50	140.40	121.82	111.87	86.68	58.20	35.96	20.87
2006.58	141.31	121.12	109.85	84.51	56.92	35.68	21.08
2006.67	142.10	120.63	108.23	82.55	55.54	35.05	20.89
2006.75	142.70	120.13	106.74	80.68	54.09	34.24	20.49
2006.83	143.34	119.38	104.91	78.65	52.53	33.32	20.05
2006.92	143.10	118.08	103.05	77.21	52.03	33.55	20.53
2007.00	141.97	116.71	102.37	77.69	53.28	34.90	21.59
2007.08	139.77	116.00	103.80	80.07	55.16	35.98	22.26
2007.17	137.87	116.79	106.61	81.96	55.11	35.44	22.34
2007.25	136.76	118.97	110.25	83.59	54.30	34.47	22.52
2007.33	136.44	122.03	114.48	85.39	53.06	32.61	21.91
2007.42	136.35	125.01	119.03	88.27	52.85	30.87	20.74
2007.50	136.30	127.57	123.21	91.53	53.63	29.95	19.72
2007.58	135.59	129.32	126.84	94.94	55.29	30.01	19.19
2007.67	135.14	130.98	130.09	98.22	57.40	30.86	19.34
2007.75	134.86	132.66	133.05	101.20	59.54	32.11	19.99
2007.83	134.72	134.33	135.88	103.97	61.58	33.37	20.71
2007.92	135.31	136.41	138.62	106.21	62.76	33.70	20.71
2008.00	136.69	138.29	140.04	106.52	62.30	33.16	20.44
2008.08	138.98	139.53	139.36	104.91	61.26	33.02	20.63
2008.17	141.11	139.75	137.91	103.98	61.88	34.17	21.09
2008.25	142.77	138.86	135.76	103.14	62.86	35.32	21.18
2008.33	143.48	136.77	132.52	101.62	63.71	36.72	21.59
2008.42	144.15	134.87	129.02	99.09	63.53	37.86	22.42
2008.50	145.01	133.77	126.21	96.49	62.62	38.31	23.10
2008.58	145.98	133.12	123.99	93.98	61.15	38.02	23.35
2008.67	146.79	132.49	122.16	91.74	59.42	37.12	23.06
2008.75	147.29	131.65	120.37	89.62	57.63	35.95	22.44
2008.83	147.69	130.88	118.71	87.67	56.02	34.97	21.99
2008.92	147.34	129.53	116.79	86.21	55.65	35.36	22.43
2009.00	146.10	127.59	115.12	85.99	56.82	36.79	23.27
2009.08	144.16	125.62	114.44	87.19	58.83	38.14	23.60
2009.17	142.50	125.12	115.66	89.41	60.46	38.42	23.17

2009.25	141.09	126.24	118.88	92.37	61.49	37.93	22.57
2009.33	140.23	128.41	123.03	95.79	62.56	37.33	21.81
2009.42	138.97	129.96	126.83	99.56	64.55	37.51	21.28
2009.50	137.34	130.12	129.11	102.56	66.74	38.38	21.25
2009.58	136.19	130.25	130.77	104.92	68.69	39.42	21.55
2009.67	135.23	130.45	132.02	106.74	70.33	40.46	22.03
2009.75	134.72	130.85	133.17	108.25	71.70	41.43	22.61
2009.83	134.16	130.94	133.77	109.32	72.99	42.50	23.24
2009.92	133.97	131.21	134.55	110.56	74.04	42.90	23.18
2010.00	134.18	132.28	136.44	112.27	74.61	42.50	22.66
2010.08	135.17	134.71	139.01	113.01	73.39	40.93	21.87
2010.17	136.68	136.86	139.48	110.54	70.02	39.06	21.72
2010.25	138.64	138.15	138.14	106.98	67.00	38.13	22.15
2010.33	140.91	139.18	136.18	102.84	63.56	37.00	22.62
2010.42	143.55	140.89	135.26	99.40	59.82	34.99	22.37
2010.50	145.58	143.11	135.97	97.81	57.10	32.93	21.62
2010.58	146.80	144.74	136.88	97.21	55.51	31.39	20.74
2010.67	147.96	146.23	137.87	97.17	54.78	30.57	20.15
2010.75	148.83	147.66	138.98	97.42	54.46	30.12	19.78
2010.83	149.93	148.99	140.27	98.16	54.56	30.03	19.82
2010.92	151.09	150.50	141.46	98.25	53.96	29.67	19.95
2011.00	152.44	151.70	141.63	97.49	53.31	29.78	20.55
2011.08	152.95	151.45	141.08	97.67	54.50	31.34	21.84
2011.17	152.60	150.47	141.09	99.50	56.91	32.93	22.42
2011.25	151.78	149.30	141.19	101.49	59.22	34.05	22.40
2011.33	150.73	148.26	142.00	104.66	62.77	35.93	22.59
2011.42	148.87	146.18	141.59	106.72	65.95	38.19	23.31
2011.50	147.94	144.59	140.56	107.32	67.80	39.98	24.21
2011.58	147.38	143.46	139.73	107.45	68.82	41.21	25.06
2011.67	146.59	142.23	138.89	107.25	69.15	41.78	25.58
2011.75	145.96	141.13	138.10	107.04	69.36	42.13	25.94
2011.83	145.28	140.30	137.28	106.39	69.05	42.01	25.81
2011.92	144.57	139.47	136.68	106.33	69.27	41.97	25.40
2012.00	143.95	138.94	136.75	106.93	69.61	41.55	24.51
2012.08	143.67	139.25	137.17	106.56	68.31	39.95	23.21
2012.17	143.76	139.71	136.91	105.02	66.34	38.62	22.75
2012.25	144.18	140.42	136.80	103.35	64.26	37.58	22.81
2012.33	144.55	141.42	136.78	101.27	61.52	36.06	22.62
2012.42	144.73	142.74	137.29	99.67	58.83	34.12	21.99
2012.50	144.44	143.93	138.31	99.03	56.94	32.42	21.21
2012.58	144.82	145.11	139.55	99.44	56.19	31.25	20.40
2012.67	145.25	146.04	140.42	99.88	55.88	30.53	19.80
2012.75	145.70	146.72	141.12	100.43	55.93	30.18	19.33
2012.83	145.93	147.21	141.93	101.29	56.30	30.01	19.01
2012.92	146.30	147.69	142.54	101.65	56.06	29.50	18.73
2013.00	146.81	148.04	142.36	100.63	54.58	28.37	18.24
2013.08	147.17	147.20	140.12	98.06	53.12	28.14	18.40
2013.17	147.04	144.82	136.48	95.92	53.43	29.45	19.21
2013.25	145.95	141.07	132.45	94.77	55.04	31.56	20.40
2013.33	144.73	137.10	128.52	93.69	56.48	33.60	21.77
2013.42	144.19	133.32	124.26	91.94	57.21	35.08	22.82
2013.50	143.83	130.05	120.07	89.70	57.35	36.12	23.60
2013.58	143.83	127.71	116.20	86.64	56.35	36.46	24.17
2013.67	143.73	125.67	112.97	83.83	55.02	36.29	24.36

2013.75	143.36	123.80	110.20	81.28	53.51	35.71	24.20
2013.83	142.70	122.05	107.77	78.83	51.74	34.74	23.74
2013.92	141.52	120.26	105.44	76.67	50.81	35.05	24.40
2014.00	140.01	118.30	103.38	75.72	51.55	36.61	25.72
2014.08	138.05	116.59	103.16	77.29	53.47	37.73	26.22
2014.17	136.07	116.13	105.11	79.96	54.74	37.50	25.70
2014.25	134.46	116.91	107.90	82.56	55.60	36.80	24.74
2014.33	132.96	117.94	110.61	85.19	56.90	36.48	23.72
2014.42	131.55	118.94	113.36	88.12	58.92	37.20	23.52
2014.50	130.35	119.53	115.47	90.54	60.79	38.19	23.75
2014.58	128.51	119.35	116.92	92.52	62.50	39.30	24.24
2014.67	126.83	119.17	118.09	94.15	63.93	40.29	24.80
2014.75	125.50	119.10	119.03	95.48	65.21	41.27	25.44
2014.83	124.43	119.02	119.87	96.88	66.66	42.35	26.02
2014.92	123.66	119.34	121.41	98.68	67.53	42.10	25.37
2015.00	123.59	120.89	123.65	99.84	67.08	40.88	24.28
2015.08	125.05	123.25	125.07	99.50	66.01	40.06	23.76
2015.17	127.37	125.45	125.37	98.34	65.06	39.76	23.56
2015.25	130.49	127.33	124.46	96.06	63.59	39.56	23.60
2015.33	133.86	128.63	122.36	92.52	61.36	39.43	24.38
2015.42	137.01	130.33	120.98	89.21	58.62	38.49	24.75
2015.50	139.40	132.10	120.60	86.88	56.15	37.20	24.60
2015.58	141.56	133.63	120.64	85.33	54.06	35.71	23.98
2015.67	143.71	135.12	120.99	84.50	52.60	34.44	23.25
2015.75	145.52	136.38	121.52	84.09	51.52	33.39	22.53
2015.83	147.00	137.32	121.83	83.57	50.55	32.68	22.32
2015.92	148.26	137.87	121.55	83.15	50.92	33.87	23.56
2016.00	149.03	137.44	120.93	83.66	52.48	35.35	24.33
2016.08	148.63	136.27	120.48	84.56	53.82	36.15	24.55
2016.17	147.74	135.00	119.82	84.63	54.31	36.85	25.27
2016.25	146.55	133.85	119.59	84.91	54.83	37.71	26.29
2016.33	145.16	132.70	119.88	85.77	55.29	37.86	26.46
2016.42	143.68	131.23	119.60	86.35	55.78	37.96	26.35
2016.50	143.04	129.95	119.04	86.68	56.23	38.15	26.33
2016.58	142.94	129.37	118.67	86.89	56.67	38.42	26.42
2016.67	142.81	128.97	118.36	86.96	56.99	38.67	26.54
2016.75	142.62	128.58	117.88	86.79	57.08	38.75	26.56
2016.83	142.63	128.38	117.60	86.61	56.95	38.49	26.25
2016.92	142.65	127.95	117.05	85.74	55.50	36.92	25.22
2017.00	142.16	127.47	116.38	84.39	53.98	36.18	25.39
2017.08	141.12	126.68	115.76	83.59	53.36	36.09	25.64
2017.17	139.46	125.83	116.04	84.62	54.36	36.43	25.45
2017.25	137.61	125.59	117.36	86.62	55.91	36.87	25.08
2017.33	135.74	125.84	119.11	88.95	58.02	37.98	25.12
2017.42	134.10	125.96	120.61	90.98	59.85	39.01	25.30
2017.50	132.60	126.04	121.75	92.58	61.44	40.04	25.64
2017.58	130.85	125.72	122.53	93.80	62.73	41.03	26.14
2017.67	129.42	125.39	123.11	94.76	63.76	41.91	26.70
2017.75	128.07	124.97	123.55	95.62	64.77	42.84	27.39
2017.83	126.86	124.32	123.66	96.50	66.17	44.19	28.22
2017.92	125.54	123.86	124.44	98.53	68.49	45.62	28.49
2018.00	124.61	123.91	126.16	101.29	70.53	46.12	28.00
2018.08	124.63	125.35	128.95	103.95	71.85	46.41	27.99
2018.17	125.69	128.07	131.98	104.98	70.93	45.36	27.61

2018.25	127.18	130.75	133.80	103.93	68.10	43.06	26.71
2018.33	129.42	134.15	136.14	103.01	64.89	40.16	25.39
2018.42	130.94	136.81	138.71	103.16	62.66	37.75	24.18
2018.50	131.38	138.41	141.16	104.21	61.58	35.99	23.10
2018.58	131.77	139.81	143.60	105.99	61.68	35.04	22.17
2018.67	131.89	140.84	145.59	107.64	62.17	34.65	21.58
2018.75	132.31	142.09	147.52	109.22	62.91	34.74	21.33
2018.83	132.91	143.59	149.49	110.53	63.20	34.46	21.03
2018.92	133.77	145.14	150.82	110.57	62.42	33.97	21.27
2019.00	134.67	145.76	150.23	109.04	61.35	34.06	22.07
2019.08	135.44	145.77	149.09	107.83	61.06	34.35	22.33
2019.17	135.42	144.49	147.49	107.68	62.25	35.48	22.78
2019.25	134.49	142.74	146.98	109.77	65.45	37.58	23.39
2019.33	132.71	140.15	145.96	111.63	68.53	39.66	23.99
2019.42	131.11	137.53	144.00	112.32	71.07	41.73	24.68
2019.50	131.08	136.29	142.12	112.10	72.76	43.61	25.61
2019.58	131.35	135.49	140.13	110.93	73.26	44.82	26.49
2019.67	131.42	134.93	138.47	109.63	73.17	45.46	27.14
2019.75	131.20	134.07	136.91	108.32	72.62	45.46	27.42
2019.83	130.79	133.19	135.48	107.20	72.30	45.74	27.81
2019.92	130.39	132.43	134.50	106.91	72.93	46.46	27.88
2020.00	129.75	131.96	134.74	108.02	73.98	46.54	27.10
2020.08	129.60	131.98	135.24	108.47	73.68	45.74	26.44
2020.17	130.08	132.45	135.27	107.31	71.96	44.74	26.37
2020.25	130.89	132.99	134.83	105.40	69.78	43.85	26.70
2020.33	131.68	133.15	134.03	103.35	67.54	42.76	26.85
2020.42	133.26	134.42	134.58	102.42	65.82	41.66	26.82
2020.50	133.85	135.01	135.36	102.32	64.69	40.53	26.37
2020.58	133.20	134.57	135.85	102.68	64.19	39.68	25.89
2020.67	132.99	134.43	136.20	102.95	63.90	39.05	25.38
2020.75	132.81	134.41	136.51	103.16	63.70	38.61	24.98
2020.83	132.84	134.59	136.81	103.23	63.35	38.12	24.68
2020.92	132.94	134.77	136.87	102.81	62.63	37.70	24.73
2021.00	133.38	135.01	136.50	101.59	61.42	37.39	25.07
2021.08	133.08	134.29	135.51	100.94	61.55	38.01	25.55
2021.17	132.50	133.92	135.95	102.30	62.59	37.99	25.07
2021.25	132.16	133.89	137.03	104.40	63.86	37.57	24.01
2021.33	131.83	133.93	138.28	107.10	66.27	38.21	23.35
2021.42	131.15	133.71	139.05	109.30	68.82	39.66	23.46
2021.50	130.79	133.63	139.26	110.35	70.57	41.08	23.97

Table S6. Summary of linear trends fitted to time series of f-N2O (ppb)

```
tropics
     32 hPa  -3.4 ± 0.8 %/dec
     22 hPa  +0.8 ± 0.8 %/dec
     15 hPa  +3.7 ± 1.4 %/dec
     10 hPa  +5.5 ± 2.0 %/dec
    6.8 hPa  +8.6 ± 2.6 %/dec
    4.6 hPa +12.0 ± 3.8 %/dec
    3.2 hPa +12.6 ± 4.7 %/dec
NH
     32 hPa  -14.9 ± 3.0 %/dec
     22 hPa   -7.7 ± 3.2 %/dec
     15 hPa   +0.6 ± 4.0 %/dec
     10 hPa   +8.4 ± 4.9 %/dec
    6.8 hPa  +16.1 ± 4.9 %/dec
    4.6 hPa  +19.5 ± 4.6 %/dec
    3.2 hPa  +18.3 ± 5.0 %/dec
SH
     32 hPa   -3.1 ± 2.5 %/dec
     22 hPa   +4.0 ± 3.4 %/dec
     15 hPa   +7.9 ± 4.3 %/dec
     10 hPa   +7.2 ± 4.6 %/dec
    6.8 hPa   +7.2 ± 4.7 %/dec
    4.6 hPa  +11.1 ± 4.7 %/dec
    3.2 hPa  +14.5 ± 3.7 %/dec
```

Table S7. N_2O budget table

	Troposphere	Burden	Lifetime	Loss	Atm. Incr.	Total	Anthrop.
	ppb	TgN	y	TgN/y	TgN/y	TgN/y	TgN/y
PI 1850s	272	1300	124.0*	10.5**	0***	10.5	0
2000s	319.6	1528	119.3	12.8	3.7	16.5	6.0
2010s	328.5	1570	117.0	13.4	4.7	18.1	7.6

Notes.
*Pre-industrial lifetime is 6% higher than current per P2015.
**The 1850s loss is assumed to be the Pre-Industrial (natural) emissions, i.e., 272 ppb (IPCC AR6) is the 'natural' PI value, and has not changed.
***The 1850s atmospheric increase in the IPCC scenarios (~0.5 ppb/y) is not observed but is derived from the assumed anthropogenic scenario.
Lifetime decreasing 2 %/dec from 2000s to 2010s.

Table S8. Calculated monthly loss of N_2O

Year	Loss N2O	
	(kgN/s)	(TgN/mon)
2004.63	375.50	1.006
2004.71	409.50	1.061
2004.79	432.00	1.157
2004.88	424.50	1.100
2004.96	408.50	1.094
2005.04	411.40	1.102
2005.13	426.30	1.031
2005.21	439.70	1.178
2005.29	424.90	1.101
2005.38	399.00	1.069
2005.46	382.20	0.991
2005.54	383.70	1.028
2005.63	403.40	1.080
2005.71	418.70	1.085
2005.79	424.40	1.137
2005.88	417.90	1.083
2005.96	432.30	1.158
2006.04	471.00	1.262
2006.13	479.70	1.160
2006.21	447.20	1.198
2006.29	400.00	1.037
2006.38	355.70	0.953
2006.46	332.40	0.862
2006.54	333.10	0.892
2006.63	349.20	0.935
2006.71	376.70	0.976
2006.79	396.30	1.061
2006.88	404.70	1.049
2006.96	404.30	1.083
2007.04	424.70	1.138
2007.13	442.50	1.070
2007.21	454.10	1.216
2007.29	434.60	1.126
2007.38	405.40	1.086
2007.46	382.10	0.990
2007.54	380.40	1.019
2007.63	397.80	1.065
2007.71	411.50	1.067
2007.79	419.70	1.124
2007.88	411.70	1.067
2007.96	417.40	1.118

2008.04	452.90	1.213
2008.13	490.20	1.186
2008.21	483.30	1.294
2008.29	430.80	1.117
2008.38	375.30	1.005
2008.46	345.20	0.895
2008.54	337.90	0.905
2008.63	349.20	0.935
2008.71	373.10	0.967
2008.79	404.40	1.083
2008.88	414.90	1.075
2008.96	417.40	1.118
2009.04	435.40	1.166
2009.13	460.60	1.114
2009.21	448.70	1.202
2009.29	422.50	1.095
2009.38	397.40	1.064
2009.46	379.60	0.984
2009.54	372.40	0.997
2009.63	377.80	1.012
2009.71	394.40	1.022
2009.79	408.40	1.094
2009.88	420.80	1.091
2009.96	453.30	1.214
2010.04	478.10	1.281
2010.12	494.70	1.197
2010.21	466.30	1.249
2010.29	428.10	1.110
2010.37	394.00	1.055
2010.46	372.50	0.966
2010.54	374.40	1.003
2010.62	402.50	1.078
2010.71	411.40	1.066
2010.79	411.10	1.101
2010.87	403.60	1.046
2010.96	422.50	1.132
2011.04	471.30	1.262
2011.12	513.10	1.241
2011.21	510.00	1.366
2011.29	466.60	1.209
2011.37	427.70	1.146
2011.46	391.40	1.015
2011.54	376.40	1.008
2011.62	382.60	1.025

2011.71	393.80	1.021
2011.79	403.00	1.079
2011.87	415.40	1.077
2011.96	428.20	1.147
2012.04	462.50	1.239
2012.12	469.10	1.135
2012.21	456.20	1.222
2012.29	424.40	1.100
2012.37	383.90	1.028
2012.46	359.20	0.931
2012.54	360.00	0.964
2012.62	383.40	1.027
2012.71	397.30	1.030
2012.79	404.00	1.082
2012.87	391.90	1.016
2012.96	387.40	1.038
2013.04	437.20	1.171
2013.12	469.60	1.136
2013.21	460.60	1.234
2013.29	420.80	1.091
2013.37	371.20	0.994
2013.46	339.60	0.880
2013.54	340.90	0.913
2013.62	371.20	0.994
2013.71	410.30	1.063
2013.79	433.70	1.162
2013.87	430.10	1.115
2013.96	410.90	1.101
2014.04	417.50	1.118
2014.12	442.40	1.070
2014.21	460.20	1.233
2014.29	450.40	1.167
2014.37	417.00	1.117
2014.46	388.50	1.007
2014.54	380.10	1.018
2014.62	400.10	1.072
2014.71	417.30	1.082
2014.79	431.40	1.155
2014.87	432.90	1.122
2014.96	445.00	1.192
2015.04	471.90	1.264
2015.12	479.20	1.159
2015.21	458.00	1.227
2015.29	424.20	1.100

2015.37	385.40	1.032
2015.46	367.30	0.952
2015.54	370.70	0.993
2015.62	383.80	1.028
2015.71	399.90	1.037
2015.79	411.00	1.101
2015.87	407.60	1.056
2015.96	406.50	1.089
2016.04	425.10	1.139
2016.12	459.10	1.111
2016.21	480.70	1.288
2016.29	464.50	1.204
2016.37	423.90	1.135
2016.46	397.30	1.030
2016.54	392.20	1.050
2016.62	407.00	1.090
2016.71	427.80	1.109
2016.79	426.90	1.143
2016.87	421.00	1.091
2016.96	415.60	1.113
2017.04	433.40	1.161
2017.12	469.10	1.135
2017.21	486.30	1.303
2017.29	468.10	1.213
2017.37	428.10	1.147
2017.46	396.20	1.027
2017.54	391.60	1.049
2017.62	415.90	1.114
2017.71	440.50	1.142
2017.79	448.40	1.201
2017.87	447.40	1.160
2017.96	442.80	1.186
2018.04	456.40	1.222
2018.12	475.70	1.151
2018.21	464.60	1.244
2018.29	428.00	1.109
2018.37	397.90	1.066
2018.46	380.20	0.985
2018.54	377.80	1.012
2018.62	393.00	1.053
2018.71	410.20	1.063
2018.79	410.60	1.100
2018.87	405.50	1.051
2018.96	429.50	1.150

2019.04	491.80	1.317
2019.12	519.60	1.257
2019.21	515.20	1.380
2019.29	486.40	1.261
2019.37	440.50	1.180
2019.46	398.40	1.033
2019.54	384.70	1.030
2019.62	396.30	1.061
2019.71	429.90	1.114
2019.79	450.20	1.206
2019.87	451.50	1.170
2019.96	461.60	1.236
2020.04	489.10	1.310
2020.12	502.30	1.215
2020.21	495.30	1.327
2020.29	475.90	1.234
2020.37	443.00	1.187
2020.46	416.40	1.079
2020.54	405.00	1.085
2020.62	416.90	1.117
2020.71	431.30	1.118
2020.79	426.90	1.143
2020.87	419.00	1.086
2020.96	426.30	1.142
2021.04	463.10	1.240
2021.12	483.60	1.170
2021.21	471.20	1.262
2021.29	448.00	1.161
2021.37	419.50	1.124
2021.46	392.00	1.016
2021.54	381.90	1.023
2021.62	392.90	1.052
2021.71	406.90	1.055
2021.79	412.50	1.105
2021.87	424.10	1.099
2021.96	451.50	1.209

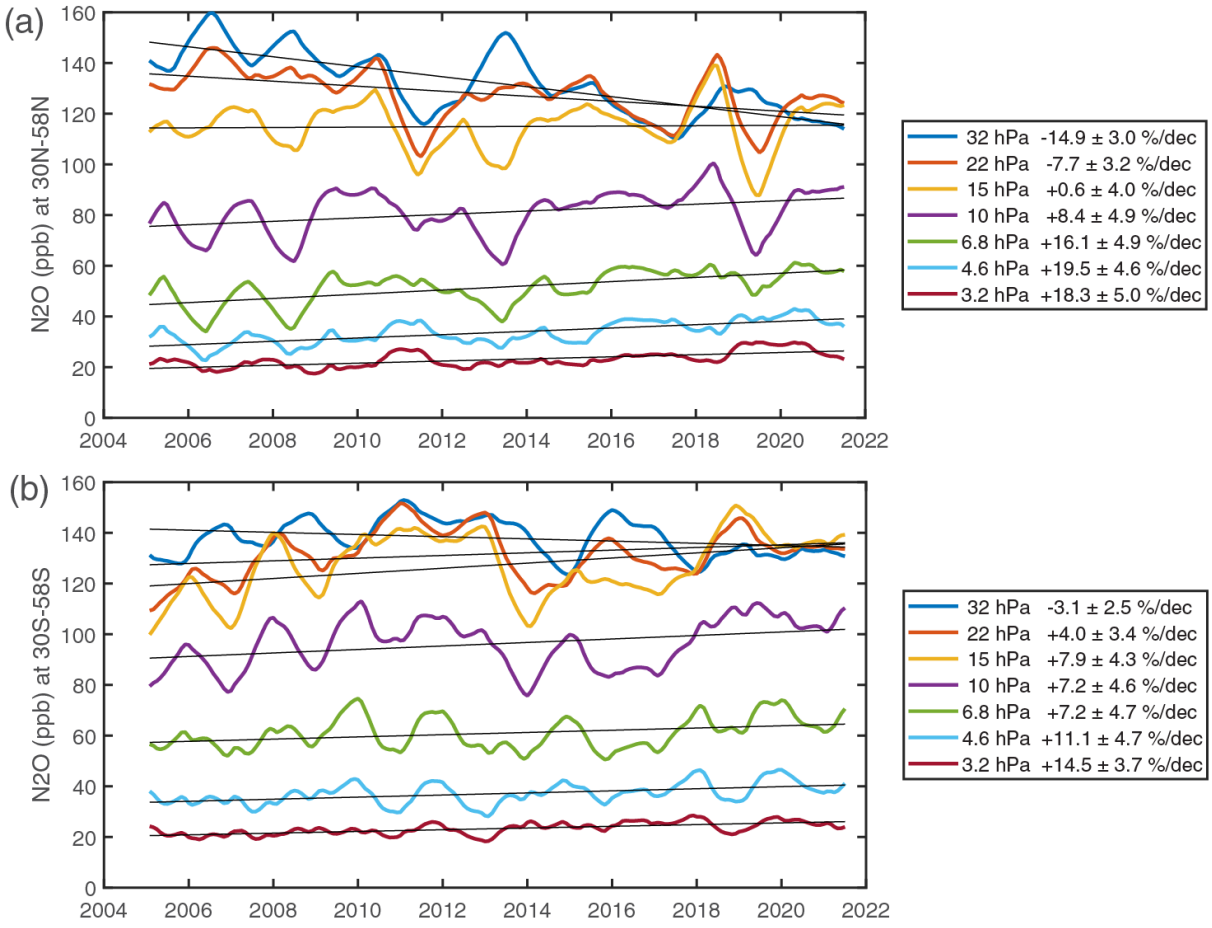

Figure S1. Time series of stratospheric f-N_2O (ppb, dry air mole fraction) at **(a)** northern mid-latitudes (30 °N – 58 °N) and **(b)** southern mid-latitudes (30 °S – 58 °S). See main text Fig. 1

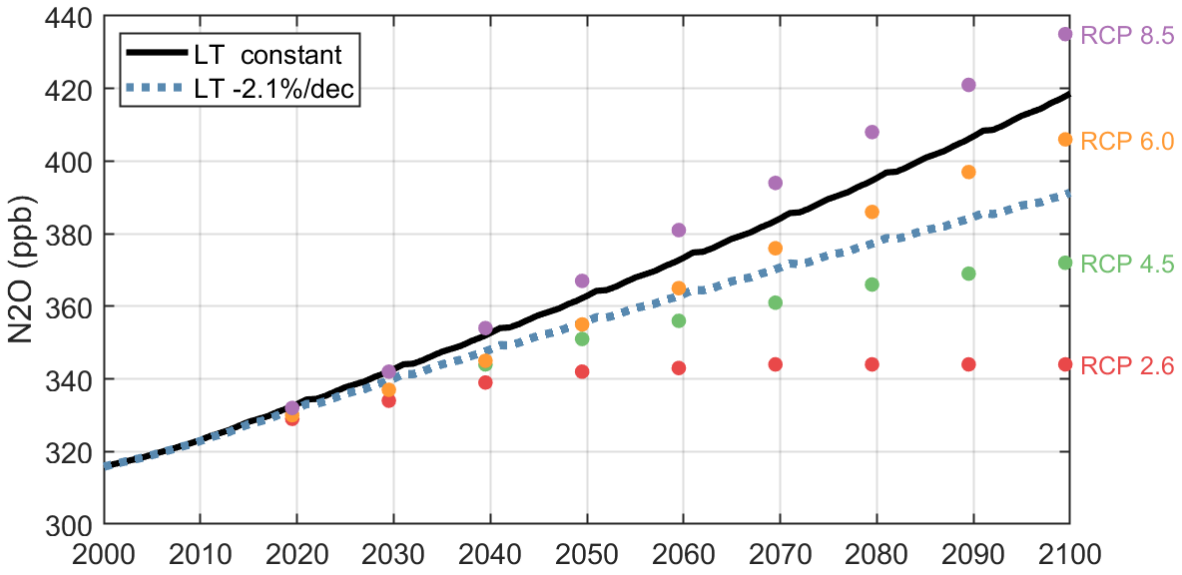

Figure S2. Projected N_2O tropospheric annual mean abundance (black line) assuming the observed 2.9 %/decade growth rate. The last observed decade 2012-2021 is used to project future decades, and hence there is a recurring interannual variability (IAV). Projected N_2O with the lifetime decreasing by 2.1 %/decade from 120 y in 2000 to 97 y in 2100 (blue dotted line) uses the emissions inferred from fixed growth scenario. The RCP scenario projections, shown for perspective, are taken from the Annex II tables of the IPCC WGI AR4 (2013).

IPCC, 2013: Annex II: Climate System Scenario Tables [Prather, M., G. Flato, P. Friedlingstein, C. Jones, J.-F. Lamarque, H. Liao and P. Rasch (eds.)]. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 1395-1445, published 31 January 2014, ISBN 978-1-107-05799-1, 2014.