https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022

Atmospheric Chemistry and Physics Discussions

Observed changes in stratospheric circulation: Decreasing lifetime of N_2O, 2005-2021

Michael J. Prather[1], Lucien Froidevaux[2], Nathaniel J. Livesey[2]

[1]Earth System Science Department, University of California Irvine; Irvine, CA 92697-3100, USA
[2]Jet Propulsion Laboratory, California Institute of Technology; Pasadena, CA 91011, USA

Correspondence to: Michael Prather (mprather@uci.edu)

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. Because N_2O abundances in the middle tropical stratosphere, where it is photochemically destroyed, increased at a faster rate than the bulk N_2O in the lower atmosphere, the lifetime is becoming shorter. The cause appears to be a more vigorous stratospheric circulation, which models predict to result from climate change. If this climate-driven circulation trend continues to 2100, then anthropogenic N_2O emissions will be removed 20% faster than current projections, and their impact on global warming and ozone depletion will be proportionately lessened. This finding is an example of a distinct negative, but relatively minor, 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 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 gases such as SF_6, but these observations run counter to the climate model projections (Karpechko et al., 2018; Abalos et al., 2021; Garney et al., 2022), in part because the comparison of models and measurements has proven difficult (Fritsch et al., 2020). The search for BDC change with AoA has

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022


Atmospheric Chemistry and Physics Discussions

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 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 lead to a shorter lifetime, an important consequence of a more vigorous BDC.

Consequences of an enhanced BDC often focus on the increased flux of stratospheric ozone (O_3) into the troposphere with subsequent increase in tropospheric O_3, see discussion in (Karpechko et al., 2018; Garney et al., 2022). Enhanced BDC also leads to shorter lifetimes for gases like nitrous oxide (N_2O) and chlorofluorocarbons (CFCs) as greater abundances are pushed higher in the tropical stratosphere where they experience greater photolytic destruction. 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). We find that the stratospheric N_2O loss rate is increasing faster 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 faster.

2 Measurements and methods

The Aura MLS observations of N_2O, O_3, and temperature (T) are taken from the Goddard Earth Sciences Data and Information Services Center in the form of Version 5 Level 3 monthly binned profiles from August 2004 through December 2021 (Lambert et al., 2021; Schwartz et al., 2021a, 2021b). All data are averaged over the 72 longitude bins. 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

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022

Atmospheric Chemistry and Physics Discussions

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 (6 evenly space log(pressure) levels per decade) is coarser than that of O_3 and T (both use the same 12 intervals per decade). The missing N_2O levels in the O_3-T grid are calculated as the square-root of the values on the adjacent levels. For pressures <0.4 hPa, where MLS V5 N_2O data are lacking, we set the abundance to 0 ppb. Polar areas outside the observations (86°-90°) are included in the weighting of the 86° profile. The dataset then covers: 43 latitudes (84°S to 84°N), 37 pressure levels (100 – 0.001 hPa), and 209 months (Aug 2004 to Dec 2021).

The observational data consists of 8,987 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 6 layers with typical tropospheric composition, but these layers do not much affect the stratospheric N_2O loss because very little of the critical radiation in these ultraviolet wavelengths reaches the troposphere. For each column we calculate the profile of N_2O loss from photolysis (J-N_2O) and reaction with the atomic oxygen radical $O(^1D)$ from the three reactions:

$$N_2O + h\nu \rightarrow N_2 + O \quad (1)$$
$$N_2O + O(^1D) \rightarrow N_2 + O_2 \quad (2)$$
$$N_2O + O(^1D) \rightarrow NO + NO \quad (3)$$

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

$$O_3 + h\nu \rightarrow O(^1D) + O_2 \quad (4)$$
$$O(^1D) + N_2 \rightarrow O(^3P) + N_2 \quad (5)$$
$$O(^1D) + O_2 \rightarrow O(^3P) + O_2 \quad (6)$$

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022


Atmospheric Chemistry and Physics Discussions

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). During the MLS Aura record used here, we believe that the solar cycle has had little impact: 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 decrease 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.

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-day years, and thus our annual losses are biased low by 0.07%. Lifetimes are budgetary time scales and calculated as a 12-month burden (TgN) divided by the 12-month loss (TgN/yr). The burden is calculated from monthly-mean, globally-averaged NOAA marine surface measurements (Dlugokencky, 2022) and using the conversion factor of 4.78 TgN/ppb (Prather et al., 2012). Given that the gridding and source files have changed since P2015, we compared the global monthly loss rates for the overlap period with P2015 (Aug 2004 – Dec 2011): the mean difference of 0.2% and rms difference of 0.6% show that both calculations are essentially identical.

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' (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

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022


Atmospheric Chemistry and Physics Discussions

The primary calculation here is the annual N_2O loss (TgN/yr) shown in Figure 1a. It was surprising in that 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±0.7 %/decade. This is larger than the +2.9±0.02 %/decade trend in the burden (Figure 1b), and results in an N_2O budgetary lifetime trend of -2.1±0.7 %/decade (Figure 1c). Our best estimate for the N_2O lifetime over the past decade (117 yr) 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 with an interannual standard deviation of 3% and 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 (3), and we calculate the production of NO (Figure 1d), which (like the N_2O loss) also shows a positive trend of +3.9±0.6 %/decade.

The cause of the lifetime trends can be a change in the photochemical loss frequency, or an increase in the abundance of N_2O. 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, and thus we focus on the tropical middle stratosphere. Photolysis (reaction 1) dominates loss here (by more than 90%) and the monthly mean J-N_2O at the equator (Figure 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 decline would reduce the loss.

The declining J-N_2O is clearly 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 of J-N_2O. For the upper stratospheric tropics (1-10 hPa), the generally observed O_3 trend of order +1.0 to +1.6 %/decade is consistent with the vertical

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022


Atmospheric Chemistry and Physics Discussions

pattern and magnitude of the J-N_2O trend. This trend reduces the N_2O loss and makes disagreement between the burden and loss trends 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 faster. 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 Figure 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 probably due to a residual negative drift of the V5 MLS data in the lower stratosphere after 2010 as shown in Figure 16 of Livesey et al. (2021). It is unlikely to be real because suppression of N_2O below 30 hPa can hardly lead to increases 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±1.2 %/decade in the central zone (10 hPa) and +12.0±2.3 %/decade near the top (4.6 hPa). These increases are used directly in our calculation and average to +6 %/decade needed to explain the trend in total N_2O loss.

Are these increases in N_2O real or an artefact of a known calibration drift in the MLS 190 GHz (mainly water vapor and N_2O) observations? The MLS Version 5 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 Figure 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 Figure 12) find similar trends in N_2O to ours

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022


Atmospheric Chemistry and Physics Discussions

(~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 Figure 3 of Prather et al., 2015), the tropics should dominate the mean value and its trend.

4 Implications

The recent WMO Ozone Assessments (Karpechko et al., 2018; Garny et al., 2022) concluded that disagreement remains regarding the direction of the BDC trend between the chemistry-climate model simulations (increasing rates) and the satellite observations (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 Figure 1f for the extra-tropics (i.e., 30°N-58°N and 30°S-58°S), then we find an almost identical pattern to that in the tropics: increasing at 7-8 %/decade versus 6 at 10 hPa; and at 14-18 %/decade versus 13 at 3.2 hPa, see Supplement Figure S1ab 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 tropics and extra-tropics. The use of an integrated quantity 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. The climate impact of N_2O emissions will drop because their decay time will fall from 110 yr to 90 yr. Note that the decay time of a pulse based on a lifetime of 117 yr is reduced by a factor of 0.94 due to chemical feedbacks (Prather, 1998; Prather et al., 2015). Overall, we will see an

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022


Atmospheric Chemistry and Physics Discussions

accelerated removal of the long-lived trace gases. The chemistry-climate model intercomparison projects should encourage the calculation of N_2O and CFC lifetimes over the 21st century as a straightforward diagnostic of changing BDC.

There is an additional wrinkle in this analysis. We find that the production of NO was not increasing as fast as the loss of N_2O, although admittedly 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 proportional much less NO (see P2015, Figure 1). This result supports a finding in 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 less than that of N_2O (approximately 2 vs. 12 %/decade); however, the abundance of NO in the tropical upper stratosphere is a balance between production, vertical transport, and photochemical loss above 40 km, and cannot be estimated from the production alone. Since this NO production is the cause of O_3 depletion by N_2O in the middle stratosphere, we may find that an enhanced BDC makes N_2O less important as an ozone-depleting gas from both lifetime changes and NO yields.

Data Availability. The raw data used here were downloaded from the sites and at the dates specified. The time series calculated here and shown in the figures are included in Tables S1-S5 in the Supplement.

Supplement. The Supplement related to this article is available online at: https://doi.org/10.5194/acp-22-.....

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

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022

Atmospheric Chemistry and Physics Discussions

Competing interests. The contact author has declared that neither they nor their co-authors have any competing interests

Acknowledgments. We thank J.L. 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 data sets used here (O_3, T, and especially N_2O).

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 was performed under contract with NASA (80NM0018D0004).

References

Abalos, M., Calvo, N., Benito-Barca, S., Garny, H., Hardiman, S. C., Lin, P., Andrews, M. B., Butchart, N., Garcia, R., Orbe, C., Saint-Martin, D., Watanabe, S., and Yoshida, K.: The Brewer–Dobson circulation in CMIP6, Atmos. Chem. Phys., 21, 13571–13591, https://doi.org/10.5194/acp-21-13571-2021, 2021.

Bernath P. F., Steffen J., Crouse J., and Boone C. D.: Sixteen-year trends in atmospheric trace gases from orbit, J. Quant. Spectrosc. Ra. 253, 107178, https://doi.org/10.1016/j.jqsrt.2020.107178, 2020.

Butchart, N. (2014), The Brewer-Dobson circulation, Rev. Geophys.,52, 157–184, doi:10.1002/2013RG000448.

Dlugokencky, E. (2022) Data from the NOAA web site, www.esrl.noaa.gov/gmd/ccgg/trends_n2o/n2o_mm_gl.txt [pulled 2022-07-26], ed.dlugokencky@noaa.gov

Fritsch, F., Garny, H., Engel, A., Bönisch, H., and Eichinger, R.: Sensitivity of age of air trends to the derivation method for non-linear increasing inert SF6, Atmos. Chem. Phys., 20, 8709–8725, https://doi.org/10.5194/acp-20-8709-2020, 2020.

Froidevaux, L., Kinnison, D. E., Wang, R., Anderson, J., and Fuller, R. A.: Evaluation of CESM1 (WACCM) free-running and specified dynamics atmospheric composition simulations using global multispecies satellite data records, Atmos. Chem. Phys., 19, 4783–4821, https://doi.org/10.5194/acp-19-4783-2019, 2019.

Froidevaux, L., Kinnison, D. E., Santee, M. L., Millán, L. F., Livesey, N. J., Read, W. G., Bardeen, C. G., Orlando, J. J., and Fuller, R. A.: Upper stratospheric ClO and HOCl trends (2005–2020): Aura Microwave Limb Sounder and model results, Atmos. Chem. Phys., 22, 4779–4799, https://doi.org/10.5194/acp-22-4779-2022, 2022.

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022

Garny, H., H. Hendon (Lead Authors), M. Abalos, G. Chiodo, A. Purich, W. Randel K. Smith and D. Thompson, Scientific Assessment of Ozone Depletion: 2022, World Meteorological Organisation Global Ozone Research and Monitoring Project-Report No. XX, WMO/UNEP Scientific Assessment of Ozone Depletion: 2022. Chapter 5: Stratospheric Ozone Changes and Climate, World Meteorological Organization, Geneva, Switzerland, 2022.

Godin-Beekmann, S., Azouz, N., Sofieva, V., Hubert, D., Petropavlovskikh, I., Effertz, P., Ancellet, G., Degenstein, D., Zawada, D., Froidevaux, L., Frith, S., Wild, J., Davis, S., Steinbrecht, W., Leblanc, T., Querel, R., Tourpali, K., Damadeo, R., Maillard-Barras, E., Stübi, R., Vigouroux, C., Arosio, C., Nedoluha, G., Boyd, I., and van Malderen, R.: Updated trends of the stratospheric ozone vertical distribution in the 60° S–60° N latitude range based on the LOTUS regression model, Atmos. Chem. Phys. Discuss. [preprint], https://doi.org/10.5194/acp-2022-137, accepted, 2022.

Karpechko, A., A. Maycock (Lead Authors), M. Abalos, J. Arblaster, H. Akiyoshi, C. Garfinkel, K. Rosenlof and M. Sigmond, Scientific Assessment of Ozone Depletion: 2018, World Meteorological Organisation Global Ozone Research and Monitoring Project-Report No. 58, WMO/UNEP Scientific Assessment of Ozone Depletion: 2018. Chapter 5: Stratospheric Ozone Changes and Climate, World Meteorological Organization, Geneva, Switzerland, 2018.

Lambert, A., Livesey, N., Read, W., and Fuller, R., MLS/Aura Level 3 Monthly Binned Nitrous Oxide (N2O) Mixing Ratio on Assorted Grids V005, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center (GES DISC), Accessed: [2022-05-06], 10.5067/Aura/MLS/DATA/3545, 2021.

Livesey, N. J., Read, W. G., Froidevaux, L., Lambert, A., Santee, M. L., Schwartz, M. J., Millán, L. F., Jarnot, R. F., Wagner, P. A., Hurst, D. F., Walker, K. A., Sheese, P. E., and Nedoluha, G. E.: Investigation and amelioration of long-term instrumental drifts in water vapor and nitrous oxide measurements from the Aura Microwave Limb Sounder (MLS) and their implications for studies of variability and trends, Atmos. Chem. Phys., 21, 15409–15430, https://doi.org/10.5194/acp-21-15409-2021, 2021.

Neu, J. L., and Plumb, R. A., Age of air in a "leaky pipe" model of stratospheric transport, J. Geophys. Res., 104(D16), 19243– 19255, doi:10.1029/1999JD900251, 1999.

Plumb, R. A., and J. D. Mahlman, The zonally-averaged transport characteristics of the GFDL general circulation/tracer model, J. Atmos. Sci.,44, 298–327, doi: 10.1175/1520-0469(1987)044%3C0298:TZATCO%3E2.0.CO;2, 1987.

Prather, M.J., Time scales in atmospheric chemistry: coupled perturbations to N2O, NOy, and O3, Science, 279, 1339-1341, 1998.

Prather, M.J., C.D. Holmes, and J. Hsu, Reactive greenhouse gas scenarios: Systematic exploration of uncertainties and the role of atmospheric chemistry, Geophys. Res. Lett., 39, L09803, 5 pp., doi:10.1029/2012GL051440, 2012.

Prather, M.J., J. Hsu, N.M. DeLuca, C.H. Jackman, L.D. Oman, A.R. Douglass, E.L. Fleming, S.E. Strahan, and S.D. Steenrod, O.A. Søvde, I.S.A. Isaksen, L. Froidevaux, and B. Funke Measuring and modeling the lifetime of nitrous oxide including its variability, J. Geophys. Res. Atmos., 120, 5693–5705. doi: 10.1002/2015JD023267, 2015.

Ruiz, Daniel J., Michael J. Prather, Susan E. Strahan, Rona L. Thompson, Lucien Froidevaux, Stephen D. Steenrod, How atmospheric chemistry and transport drive surface variability of N2O and CFC-

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022

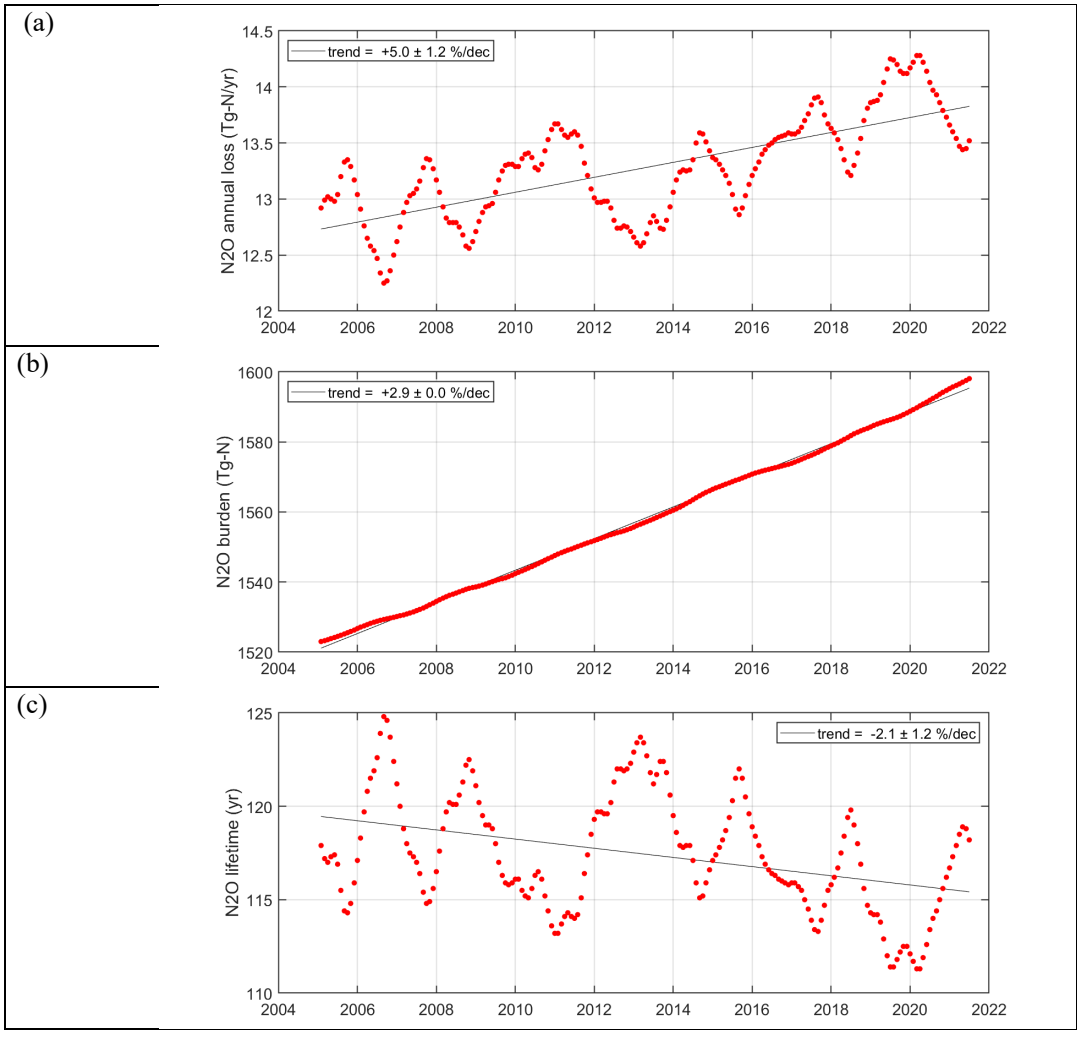
11. J. Geophys. Res.: Atmospheres, 126, e2020JD033979. https://doi.org/10.1029/2020JD033979, 2021.

Schwartz, M., Livesey, N., Read, W., and Fuller, R., MLS/Aura Level 3 Monthly Binned Temperature on Assorted Grids V005, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center (GES DISC), Accessed: [2022-07-26] [Data Access Date], 10.5067/Aura/MLS/DATA/3550, 2021a.

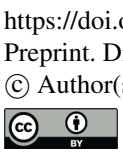
Schwartz, M., Froidevaux, L., Livesey, N., Read, W., and Fuller, R., MLS/Aura Level 3 Monthly Binned Ozone (O3) Mixing Ratio on Assorted Grids V005, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center (GES DISC), Accessed: [2022-05-06, yrs 2004 & 2021 re-pulled on 2022-07-06], 10.5067/Aura/MLS/DATA/3546, 2021b.

https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022

Atmospheric Chemistry and Physics Discussions



https://doi.org/10.5194/acp-2022-650
Preprint. Discussion started: 27 September 2022



Atmospheric Chemistry and Physics Discussions

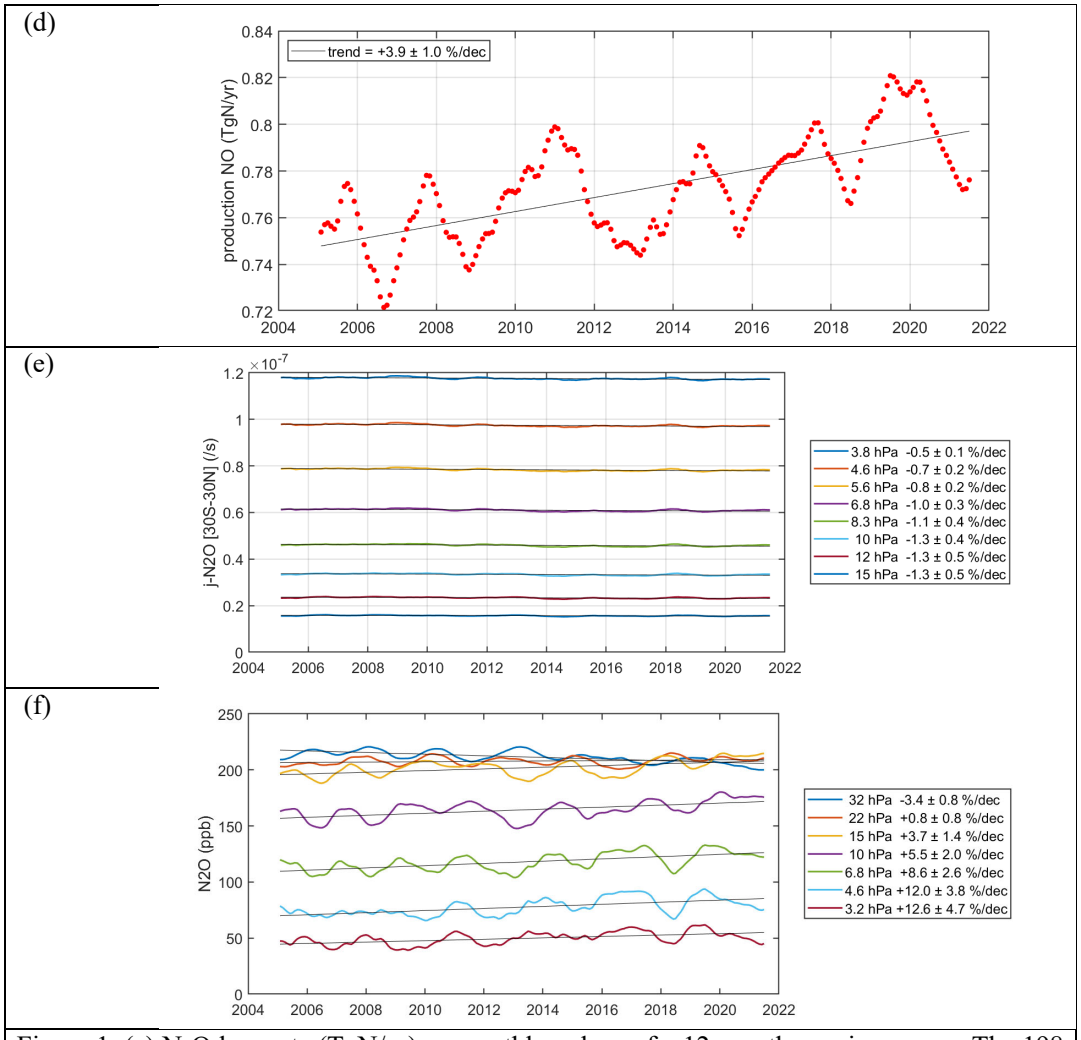


Figure 1. (a) N_2O loss rate (TgN/yr) as monthly values of a 12-month running mean. The 198 red points begin with the 12-month average of August 2004 through July 2005 plotted as February 1, 2005 with the final point being July 1, 2021 (all of 2021). The thin black trend line show the slope and uncertainty in the legend, see text. (b) N_2O global burden (TgN) based on marine surface observations. (c) N_2O lifetime derived from the burden divided by loss rate. (d) Production of NO (TgN/yr) 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.