Long-Term Variation of Greenhouse Gas N₂O Observed by MLS during 2005–2020

Xinyi Lan, Liye Zhu, and Qiangqiang Yuan

1 School of Atmospheric Sciences, Sun Yat-sen University, Zhuhai 519082, China; lanxy9@mail2.sysu.edu.cn
2 Southern Marine Science and Engineering Guangdong Laboratory (Zhuhai), Zhuhai 519082, China
3 Key Laboratory of Tropical Atmosphere-Ocean System, Ministry of Education, Zhuhai 519082, China
4 School of Geodesy and Geomatics, Wuhan University, Wuhan 430079, China; qqyuan@sgg.whu.edu.cn

* Correspondence: zhuly37@mail.sysu.edu.cn

Abstract: Nitrous oxide (N₂O) is a potent and long-lived greenhouse gas that contributes to global warming with a global warming potential (GWP) 298 times that of carbon dioxide (CO₂). In this paper, we analyzed the trend of N₂O concentration in vertical layers of the stratosphere from 2005 to 2020 using the N₂O observed from the Microwave Limb Sounder (MLS) that is on board the Aura satellite. We found that the local N₂O concentration showed a downward trend in the lower stratosphere but rose or fluctuated in the upper stratosphere. The reduction reached −5 ppb/yr at pressure levels of 31.62 hPa and 68.13 hPa, with a confidence level of over 90%. The growth was around 1–2 ppb/yr in the upper stratosphere. In addition, a concentration anomaly was observed in the tropical stratosphere in 2013. After the appearance of this anomaly, the N₂O concentration in the middle and lower layers of the tropical stratosphere was lower than before 2013. We speculated that the enhancement of the Brewer–Dobson circulation (BDC) upwelling before and after stratospheric sudden warming (SSW) is the main reason for the abnormal concentration distribution in 2013. Stratospheric N₂O has changed significantly in the past 16 years with the mutual coupling effect of BDC and SSW and such changes can have further impact on the chemical equilibrium and radiation balance in the stratosphere, as well as on the persistent climate-warming trend.

Keywords: long-term variation; stratospheric N₂O; Microwave Limb Sounder (MLS); Brewer–Dobson circulation (BDC); sudden warming of the stratosphere (SSW)

1. Introduction

As one of the most important greenhouse gases, nitrous oxide (N₂O) has a global warming potential (GWP) 298 times that of carbon dioxide (CO₂) on a per-molecule basis [1]. N₂O is mainly produced as a by-product of microbial nitrification and denitrification in soil and the ocean [1,2]. Due to the use of nitrogen-containing fertilizers in agricultural production, the tropospheric volume mixing ratio (VMR) of N₂O increased by 20%, from 270 ppb in pre-industrial times to 324 ppb in 2011 [3]. In the context of global warming, controlling N₂O is also regarded as a crucial part of controlling the greenhouse effect [4]. However, N₂O is largely ignored in emission reduction policies. Resulting from the globe’s extensive use of synthetic nitrogen fertilizer, global human-induced emissions have increased by 30% over the past four decades and exceeded all the shared socioeconomic pathways (SSPs) described in the sixth assessment report of the Intergovernmental Panel on Climate Change (IPCC) [5].

The rise of N₂O concentration brings not only the threat of global warming, but also the risk of stratospheric ozone depletion. The transport of N₂O from the troposphere to the stratosphere and its distribution in the stratosphere are dominated by Brewer–Dobson circulation (BDC) [6–9]. In addition, sudden stratospheric warming (SSW) can also significantly change the N₂O distribution [10–13]. There are increasing simulation
results showing that with the increase of greenhouse gas emissions, transport from the troposphere to the stratosphere in BDC may increase [14–17], and the frequency of SSW may also increase [18]. This indicates that there may be greater uncertainty in the distribution of greenhouse gases in the stratosphere in the future, which in turn affects BDC and SSW.

The current studies on the average level and emission estimation of N₂O are relatively limited, and research on the distribution of stratospheric N₂O is also lacking. We believe that it is necessary to analyze and display the change in the distribution of N₂O in the stratosphere in recent years and sort out the possible links of various elements. We wish to fill this research gap and provide reference for further research.

2. Data and Methods

2.1. Data Sources

N₂O is a trace gas that accounts for a very small proportion in the atmosphere, hiding in many other gases of greater concentration. Therefore, it is necessary to use measurements with great sensitivity and extreme specificity, instead of large-scale measurements, to determine N₂O concentration. Two observational projects, Halogen Occultation Experiment (HALOE) and Microwave Limb Sounder (MLS), greatly improve the understanding of stratospheric chemistry and the stratospheric climate related to N₂O. In 1993, HALOE, one of the highly instrumented experiments on NASA’s Upper Atmosphere Research Satellite (UARS), acquired observational results of trace chemical components including N₂O in the stratosphere and mesosphere using gas filter correlation radiometry [19]. A decade later, in 2004, the Earth Observing System Microwave Limb Sounder (MLS) began full scientific operation on 13 August and observed N₂O concentration in stratosphere [20].

Among the available satellite observations, the observation from MLS has more applicability than that derived from HALOE, since MLS products have good vertical resolution in the stratosphere and MLS takes measurements every 1.5° in latitude and passes through the equator 14 times a day, whereas HALOE conducts only a few tropical measurements per month.

MLS on board the Aura satellite was launched in 2004. It orbits 705 km (438 miles) above the earth, with a 98.8 min period and 240 times scanning per orbit. It crosses the equator at 1:45 p.m. [7]. The spatial coverage of the MLS standard data product is 82° N–82° S. Each observation is 1.5° or ~165 km along the orbit track (about 15 tracks per day), and the vertical resolution is 4–6 km [21]. The standard products of N₂O are taken from 190 GHz and 640 GHz retrieval [22,23]. The data used in this study are from MLS Level 2 N₂O Mixing Ratio V005 (retrieved from Earth Data website: https://disc.gsfc.nasa.gov/datasets/, last accessed on 6 May 2021). The horizontal coverage of the data is from 81.75° S to 81.75° N, and the vertical resolution is 4 – 6 km with a coverage of 68.13 hPa–0.46 hPa (14 layers in total). We selected MLS data from 2005 to 2020 and filtered the data by excluding observations with (1) negative precision, (2) an odd status field, (3) a quality field of less than 1, and (4) a convergence field greater than 2, as suggested in Livesey et al. [21].

2.2. Trend Analysis

The purpose of this study is to reveal the long-term changes in N₂O concentration and the global distribution. We analyzed the temporal variation trend of each grid at each pressure level and presented the changing rate per year in color (Figures 1 and 2).

We first averaged the MLS observations to a gridded map with a horizontal resolution of 1.5° × 1.5° for each month and each pressure level C_{i,j,l,y,m}. The annual average concentration in each grid C_{i,j,l}(y) is calculated as:

$$C_{i,j,l}(y) = \frac{1}{12} \sum_{m=1}^{12} C_{i,j,l,y,m},$$  

(1)

where i is the index of latitude, j is the index of longitude, l is the index of pressure level, y is the year, and m is the month.
The annual mean time series corresponding to spatial grid cells were then processed by univariate linear regression, and the fitting slope \( K_{i,j} \) was taken as the concentration changing rate of the spatial grid. We only show on the graph the annual rate \( (K_{i,j}) \) of grid cells with a trend analysis confidence level greater than 90\% (Figures 1a,b and 2a–d).

The zonal mean \( C_{i,j,y,m} \) of the latitudes is calculated as:

\[
C_{i,j,y,m} = \frac{1}{n_j} \sum_{j=1}^{n_j} C_{i,j,i,y,m},
\]

where \( n_j \) is the number of grids in longitude. The time series of the zonal means are displayed in Figures 1c,d and 2e–h.

In addition, the regional average \( RA \) and anomaly \( RA' \) are given as follows:

\[
RA_{i,j,y,m} = \frac{1}{n_j} \sum_{i=1}^{n_i} \cos \phi_i \frac{\sum_{j=1}^{n_j} C_{i,j,i,y,m}}{n_j},
\]

\[
RA'_{i,j,y,m} = RA_{i,j,y,m} - \frac{1}{16} \sum_{y=2005}^{2020} \frac{1}{12} \sum_{m=1}^{12} RA_{i,j,y,m},
\]

where \( \phi \) is the latitude. We show the results of different regions in Figure 3.

**Figure 1.** (a,b) Annual rate of \( N_2O \) concentration at pressure levels of 3.16 hPa and 4.64 hPa from 2005 to 2020, respectively; (c,d) zonal mean time series and trend analysis of \( N_2O \) concentration at pressure levels of 3.16 hPa and 4.64 hPa from 2005 to 2020, respectively. Only trends with a confidence level greater than 90\% are shown on the graph.
Figure 2. (a–d) Annual rate of $N_2O$ concentration at pressure levels of 21.54 hPa, 31.62 hPa, 46.42 hPa, and 68.13 hPa from 2005 to 2020, respectively; (e–h) zonal mean time series and trend analysis of $N_2O$ concentration at pressure levels of 21.54 hPa, 31.62 hPa, 46.42 hPa, and 68.13 hPa from 2005 to 2020, respectively. Only trends with a confidence level greater than 90% are shown on the graph.
Figure 3. Temporal variation of N$_2$O horizontal regional average concentration (left) and its anomaly (right) at all vertical pressure levels from 2005 to 2020 over the region of latitude of 23.5°S–23.5°N (a,b) and of 81.75°S–81.75°N (c,d).

3. Results

3.1. The Long-Term Variation Trend of N$_2$O at Each Layer

We found that the N$_2$O concentration showed a downward trend in the lower stratosphere but rose or fluctuated in the upper stratosphere. The increase in zonal mean of N$_2$O concentration mainly depended on the latitude. This trend distribution was similar to the zonal distribution of long-term average N$_2$O concentration in each layer (Figure A1).

3.1.1. Upper Stratosphere Trends

Overall, the global N$_2$O concentration showed an upward trend at the pressure level of 14.68 hPa–0.46 hPa. Significant growth occurred at pressure levels of 4.64 hPa and 3.16 hPa. The annual rate at the pressure level of 3.16 hPa was around 1–2 ppb/yr between 20°S and 20°N (Figure 1a,b). The annual rate of zonal average of N$_2$O concentration varied by latitude, with a relatively larger increase at the 15°S and 15°N bands than at the 45°S and 45°N bands. The annual rate at the pressure level of 4.64 hPa (Figure 1c,d) was between 1–3 ppb/yr, which is similar to that of 3.16 hPa. On the other hand, the growth at other layers (14.68 hPa, 10 hPa, 6.81 hPa, 2.15 hPa, 1.47 hPa, 1 hPa, 0.68 hPa, and 0.46 hPa) was not significant.

Between 2012 and 2013, the tropical N$_2$O concentration at 3.16 hPa and 4.64 hPa increased sharply, which could be related to the energetic particle precipitation (EPP) and SSW [6,24]. EPP initiates a chemical reaction to form an N$_2$O source in the upper atmosphere. When an SSW event occurs, upper atmospheric N$_2$O can be transmitted downward to the upper stratosphere.

3.1.2. Lower Stratosphere Trends

In the lower level of the stratosphere (68.13 hPa–21.54 hPa), we observed a downward trend of N$_2$O concentration in a large area of the globe (Figure 2). The annual rate of each layer displayed a zonal distribution. The smaller values, between −2 and −5 ppb/yr, appeared on the layers with pressure levels of 31.62 hPa and 68.13 hPa (Figure 2b,d). The distribution of the annual rate at the pressure level of 68.13 hPa was symmetrical, whereas the distributions on the other three layers were asymmetric, with a greater decrease in the northern hemisphere. The sharpest drop band was at the equator at the pressure level
of 68.13 hPa, reaching −5 ppb/yr. At 46.42 hPa, significant negative values were found in the high latitudes of the northern hemisphere and were between −1 and −4 ppb/yr (Figure 2c). The negative annual rate band at 21.54 hPa was comparatively narrow, lying between 15°N and 30°N, and the values range from −2 to −3 ppb/yr. There is no obvious decline in the southern hemisphere at the pressure levels of 21.54 hPa.

3.2. Temporal Variation of Vertical Profile

For the concentration zonal mean anomaly that appeared around 2013 (Figures 1c,d and 2f,h), we calculated the monthly average concentrations over the region of 23.5°S−23.5°N (Figure 3a,b) and 81.75°S−81.75°N (Figure 3c,d), which showed obvious trend characteristics in spatial distribution (Figures 1 and 2). To observe the change in regional average values throughout this period, Figure 3 shows the temporal variation of zonal means averaged for two latitude bands at all vertical pressure levels, with pressure as the ordinate axis and time as the abscissa.

Before 2013, the N₂O level in the middle and lower stratosphere maintained a stable quasi biennial fluctuation. Compared with the regional average of 81.75°S−81.75°N, the fluctuation amplitude of the regional average of 23.5°S−23.5°N was greater. We think such characteristics may be related to the stronger transport between the upper troposphere and the lower stratosphere in tropical stratosphere: BDC and quasi biennial oscillation (QBO) dominated the composition distribution of the tropical stratosphere to some extent [6–9,25–29]. BDC made the influence of QBO in the tropical troposphere spread to the stratosphere [30]. The photochemical process forced the fluctuation of trace gas to keep pace with the annual change in zonal average residual vertical velocity [30].

In January 2013, the regional average of N₂O level in the middle and lower stratosphere dropped sharply suddenly. The regional average of 23.5°S−23.5°N decreased more: from 329 ppb to 290 ppb at the pressure level of 68.13 hPa, and from 253 ppb to 238 ppb at the pressure level of 31.62 hPa. The regional average of 46.42 hPa and 68.13 hPa also decreased in 2018, but the decrease of 10 ppb from February to July of 2018 was smaller compared with the 20 ppb decrease from December 2012 to April 2013. Although the concentrations in the middle and lower stratosphere rebounded after 2018, the level was still lower than the average level before 2012. Taking the regional average of 23.5°S−23.5°N as an example, the average level was 272 ppb after December 2012, which is much lower than the 293 ppb before. The sharp decline from December 2012 to January 2013 and from February to March 2018 may be related to the impact of SSW events during this period [6,13].

3.3. Mutation in 2013

From the N₂O concentration trend in the lower stratosphere in Figure 2 and the vertical profile anomaly shown in Figure 3 we found that the concentration level in the lower tropical stratosphere decreased significantly from December 2012 to February 2013. This significant decrease mainly occurred in tropical regions, where it had the highest N₂O concentrations (Figure 4).

In the vertical aspect, significant changes were also found. Figure 5 shows the vertical profile of the monthly zonal mean concentration of N₂O from September 2012 to May 2013. The concentration isoline of the vertical profile of the tropical stratosphere changed into an asymmetric structure from December 2012 to February 2013. The high center (highest concentration point of each layer) moved south consistently with time, especially at lower pressure levels, and the isolines in the lower middle latitudes were dense in December 2012 and sparse in January 2013. The lower concentration isoline (<225 ppb) extended upward, whereas the higher concentration isoline (>250 ppb) moved downward. This changed distribution lasted from January to May 2013.
Figure 4. Monthly average N$_2$O concentration from December 2011 to February 2012 (a–c) and from December 2012 to February 2013 (d–f) at 68.13 hPa.

Figure 5. (a–i) Vertical profile of monthly zonal mean of N$_2$O concentration from September 2012 to May 2013.

This could have been caused by the abnormal enhancement of residual circulation in BDC during the SSW event [12]. The European Centre for Medium-Range Weather Forecasts (ECMWF) global atmospheric reanalysis data (ERA-Interim) show that there was a strong upward movement at the height of 10 hPa between the occurrence of SSW in 2013 [12]. SSW is generated by the interaction between zonal mean circulation and planetary waves [31]. Large-amplitude planetary waves (PW) dominate in the middle atmosphere in winter, which is the time period when SSW appeared in 2013 [32,33]. The large planetary wave activity before and after the SSW strengthens the vertical component.
of BDC residual circulation. We believe that the SSW event in 2013 led to the abnormal concentration distribution in the tropical stratosphere to a certain extent.

4. Discussion

\(N_2O\), as a stable trace gas, is often used in stratospheric dynamics studies. It has a long lifetime of \(116 \pm 9\) years [34]; thus, its concentration changes in the stratosphere are most likely to coincide with possible long-term trends in stratospheric air movement. However, due to the long time scale of BDC and the discrepancies in vertical velocities between different reanalysis products, it is difficult to obtain a statistically significant trend of BDC over 16 years, and there is a lack of direct and statistically significant observational evidence of BDC intensity trends (Figures A2 and A3) [35,36]. Although it is difficult to quantitatively analyze the contribution of BDC upwelling intensity to stratospheric \(N_2O\) change in the past 16 years, its sudden effect is obvious: A sudden increase in BDC in the short term during an SSW event may also lead to a sudden change in the gas concentration distribution, which can have impact on the concentration distribution over a longer period of time (Figure 3).

Although the specific physical and chemical mechanism of the changing \(N_2O\) trend in the past 16 years remains to be analyzed, it can be confirmed that stratospheric \(N_2O\) changed significantly in the past 16 years under the mutual coupling effect of BDC and SSW, and such changes will affect the chemical equilibrium and radiation balance in the stratosphere. Based on the interaction between greenhouse gases and stratospheric circulation and extreme events, the change in stratospheric greenhouse gases is likely to be more uncertain in terms of the persistent climate-warming trend.

5. Conclusions

We found that the \(N_2O\) concentration showed a downward trend in the lower stratosphere but rose or fluctuated in the upper stratosphere. The reduction reached \(-5\) ppb/yr at pressure levels of 31.62 hPa and 68.13 hPa, with a confidence level of over 90%. The growth was around 1–2 ppb/yr in the upper stratosphere. In addition, the annual rate isopleth of each layer practically parallel to the latitude and the annual changing rate of \(N_2O\) concentration in adjacent areas were relatively consistent. The belt with the large absolute value of annual rate appeared in the equator and northern hemisphere. From the vertical profile, before 2013, the \(N_2O\) level in the middle and lower stratosphere maintained a stable quasi biennial fluctuation, and the fluctuation of different layers had great synchronization.

Furthermore, there was a large concentration anomaly in the tropical stratosphere in early 2013. After this anomaly, the \(N_2O\) concentration in the middle and lower layers of the tropical stratosphere was lower than before 2013. The horizontal averaged \(N_2O\) concentration of 23.5°S–23.5°N was 272 ppb after December 2012, which is far lower than the value of 293 ppb before. It is speculated that the enhancement of BDC upwelling before and after SSW is the main factor that led to the abnormal concentration distribution in 2013 [12].

There was a high correlation between the local stratospheric \(N_2O\) concentration and the BDC tropical upwelling, but it was difficult to obtain the quantitative contribution of BDC to the trend of \(N_2O\) concentration over 16 years based on the existing observational data. The physical and chemical driving factors of the \(N_2O\) concentration trend remain to be analyzed, but it is worth noting that the \(N_2O\) concentration distribution indeed changed significantly under the combined influence of SSW and BDC. Such changes will have further impact on the chemical equilibrium and radiation balance of the stratosphere, and may influence the global warming trend.

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Data Availability Statement: MLS Level 2 N\textsubscript{2}O Mixing Ratio V005 datasets were analyzed in this study. These data can be found here: https://disc.gsfc.nasa.gov/datasets/ (accessed on 26 June 2021). The analyzed dataset generated in this study can be found here: http://www.satdatafresh.com/N2O_MLS.html. ERA-5 datasets can be found at: https://cds.climate.copernicus.eu/cdsapp#!/home (accessed on 8 November 2021).

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A. Horizontal Distribution of N\textsubscript{2}O Concentration over 16 Years

The N\textsubscript{2}O concentration in the stratosphere decreased from the lower layer to the upper layer, from the equator to the poles, and the distribution was symmetrical in the northern and southern hemispheres. The concentration gradient in the middle and lower latitudes was basically perpendicular to the latitude line; that is, the concentration isoline was approximately parallel to the latitude, and the low-level gradient was greater than the high-level gradient. As the height decreased, the concentration isoline intersected the weft in the high-latitude area.

![Figure A1. Average N\textsubscript{2}O concentration distribution of each layer from 2005 to 2019: (a) 1 hPa; (b) 2.15 hPa; (c) 4.64 hPa; (d) 10 hPa; (e) 46.42 hPa; (f) 68.13 hPa.](image)

Appendix B. Tropical Upwelling Mass Flux

We calculated tropical upwelling mass flux as a metric to quantify the intensity of deep-branch BDC using the wind field and temperature data from ERA5 provided by the European Centre for Medium-Range Weather Forecasts (ECMWF). The results show that the mass flux was highly correlated with the N\textsubscript{2}O concentration at each layer (Figure A2). The trend analysis of the tropical upwelling associated with BDC over the past 16 years

| Figure A1. Average N\textsubscript{2}O concentration distribution of each layer from 2005 to 2019: (a) 1 hPa; (b) 2.15 hPa; (c) 4.64 hPa; (d) 10 hPa; (e) 46.42 hPa; (f) 68.13 hPa. | Figure A2. Tropical upwelling mass flux as a metric to quantify the intensity of deep-branch BDC over the past 16 years.
was not statistically significant (the confidence level was only 25%, Figure A3). Thus, there is insufficient evidence that the intensity of deep-branch BDC caused the trend of N$_2$O concentration from 2005 to 2020.

BDC can be described by using the transformed Eulerian mean (TEM) residual circulation as its proxy [37,38]. In the TEM framework, the residual-mean meridional circulation ($v^*, w^*$) is defined as:

$$\begin{align*}
v^* &= v - \rho_0^{-1} \frac{\partial}{\partial z} \left( \rho_0 (\psi' \theta') \right) \\
w^* &= \bar{w} + \frac{1}{a \cos \phi} \frac{\partial}{\partial \phi} \left( \cos \phi (\psi' \bar{v}) \right)
\end{align*}$$

(A1)  
(A2)

The integrated tropical upwelling mass flux calculated over all tropical latitudes where the residual-mean vertical velocities ($w^*$) of 70 hPa pressure level are positive are regarded as a metric for quantifying the intensity of deep-branch BDC [39,40].

![Figure A2](image-url)

**Figure A2.** The correlation coefficient (R value) of the monthly average tropical upwelling mass flux at 70 hPa to the local monthly average N$_2$O concentration at (a) 1 hPa, (b) 10 hPa, (c) 21.54 hPa, (d) 31.62 hPa, (e) 46.42 hPa, and (f) 68.13 hPa. Only correlation coefficients with a confidence of more than 90% are shown on the graph.
\[ v^* = \bar{v} - \rho_0^{-1} \partial_z (\rho_0 \bar{v} \bar{\theta} \bar{z}) \] (A1)

\[ w^* = \bar{w} - \rho_0^{-1} \partial_z (\rho_0 \bar{w} \bar{\theta} \bar{z}) \] (A2)

\[ A3 \]

**Figure A3.** (a): Annual average tropical upward mass flux at 70 hPa; (b) mean residual vertical velocities \( w^* \) at 70 hPa from ERA5 for 2005–2020.

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