A PRELIMINARY REPORT ON THE NUMERICAL SIMULATION OF THE THREE-DIMENSIONAL STRUCTURE AND VARIABILITY OF ATMOSPHERIC N_2O

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Abstract. A numerical simulation of atmospheric N_2O using the GFDL 3-D tracer model with a small uniform surface source (15 Mton yr^{-1}) and stratospheric destruction (150 yr lifetime) has been run to a state near transport and chemical statistical equilibrium. The resulting N_2O tropospheric distribution is relatively uniform with a slight excess in the Southern Hemisphere. In the model stratosphere there is a sharp poleward decrease in N_2O mixing ratio away from high values in the tropics with pronounced winter minimums at 50°S and 60°N. Even with the small uniform surface source, relative standard deviations of N_2O in the surface layer range from 0.1% to 0.8%, well within the range of recent measurements. Additional experiments suggest that motions acting upon N_2O accumulation in the source region boundary layer and upon the mixing ratio gradient between the troposphere and lower stratosphere are the major sources of tropospheric N_2O variability.

Introduction and Model Description

The major goals of our study are: the identification of the dynamical processes controlling the transport of a tracer with a surface source and stratospheric sink; the analysis of space and time variability of a long-lived trace gas; the generation of a consistent N_2O field to be used as input for a 3-D numerical simulation of NO_y; and a reliable estimate of the distribution and size of tropospheric sources and sinks of N_2O. In this preliminary report we discuss, for the equilibrated uniform surface source calculation, zonal mean cross sections of N_2O mixing ratio and relative standard deviation for winter and summer, as well as one year time series from representative surface boxes and their relative standard deviations. Preliminary results from two other experiments, one with no surface source and one with a highly non-uniform source, are also discussed.

The basic tool for this study is the GFDL 3-D tracer model which solves numerically the continuity equation for the mixing ratio of N_2O (Mahlman, 1973). The required input data is generated by a GFDL general circulation model. The grid structure utilizes 11 terrain following (sigma) surfaces in the vertical with standard heights of 31.4, 22.3, 18.8, 15.5, 12.0, 8.7, 5.5, 3.1, 1.5, 0.5 and 0.1 km. The horizontal grid size is approximately 265 km. Discussions of the 3-D tracer model and references to relevant features of the general circulation model are presented in Mahlman and Moxim (1978).

The only destruction occurs in the top two layers of the model and has a destruction time (total atmospheric mass of N_2O divided by integrated destruction rate) of about 150 years. The N_2O volume mixing ratio (R_{N_2O}) destruction rate used in the continuity equation, "SINK", is given by

$$\text{"SINK"} = C[\, J4 + n(O(^1D)) \cdot K5]\, R_{N_2O} \qquad (1)$$

where J4, the daily average N_2O dissociation rate coefficient and $n(O(^1D))$, the daily average metastable atomic oxygen number density, are evaluated at the upper two sigma surfaces. The rate coefficient for $O(^1D)$-oxidation of N_2O, K5, is taken from the compilation of Hampson and Garvin (1975). J4 is calculated using the temperature-independent absorption coefficients measured by Johnston and Selwyn (1975), the solar flux data from the compilations of Ackerman (1971) and Simon (1974), the O_2 and O_3 absorption coefficients from Ackerman (1971), and observed annual zonal mean ozone profiles. The daily average number density of $O(^1D)$ was calculated from

$$n(O(^1D)) = \frac{J2 \cdot R_{O_3}}{K6}$$

where the daily average O_3 dissociation rate coefficient for λ <3150Å, J2, was evaluated in the same manner as J4 using the quantum yield for $O(^1D)$ previously used by Levy (1974). The rate coefficient for the quenching of $O(^1D)$ by air, K6, is taken from the compilation of Hampson and Garvin (1975). The term C in Eq. 1 is a factor which compensates for the error introduced by assuming that the value of Eq. 1 at the sigma surface is the mean for the whole layer.

The nature, size, and distribution of tropospheric N_2O sources and even the existence of tropospheric N_2O sinks were all uncertain when this calculation was begun in 1976 and are still so in 1978 (see e.g., McElroy, et al., 1976; Hahn and Junge, 1977; Cicerone et al., 1978). However, the necessity for a net tropospheric source to balance stratospheric destruction is well established. A simple approach for estimating the magnitude of sources and sinks was presented by Junge (1974) who related the tropospheric residence time of a trace gas to its atmospheric variability. Based on a series of tropospheric measurements made in Germany, Junge estimated for N_2O a temporal relative standard deviation of 8% which, using the Junge relationship, gave a residence time of 2 years. Based on estimated source strengths, Junge found an upper limit of 8 years for the N_2O residence time. While the short Junge residence time and large relative standard deviation appeared to require large tropospheric sources and sinks, Weiss's measured relative standard

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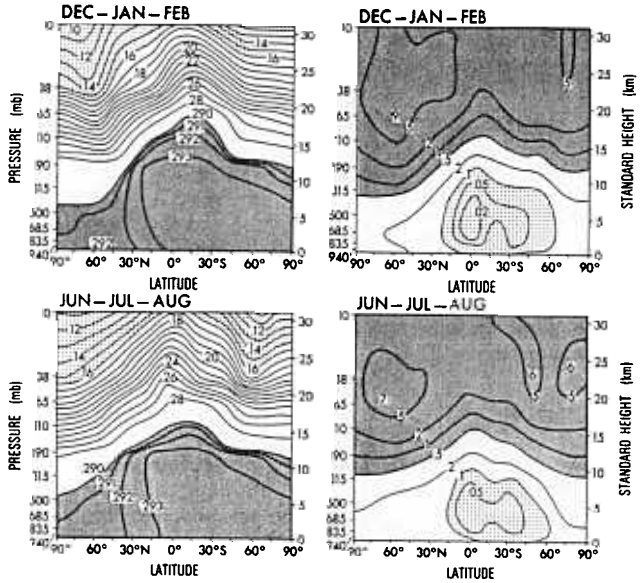

Fig. 1. On the left are winter and summer cross sections of zonal mean N_2O mixing ratios. Note smaller contour interval in the dark shaded area. On the right are winter and summer zonal mean cross sections of N_2O longitudinal relative standard deviation (V_λ). $V_\lambda = 100 \cdot (\overline{R^{*2}}^\lambda)^{1/2} / \overline{R}^\lambda$ where $(\bar{\ })^\lambda$ represents a zonal average and $(\ )^*$ is a deviation from it. Values of V_λ are not contoured in the surface layer due to resolution limitation.

deviation of 0.5% (private communication, 1976) indicated a much lower variability. Its Junge residence time of 28 years was still much shorter than the stratospheric destruction time, but no reasonable tropospheric destruction mechanism had even been proposed in 1976. We also knew from previous 3-D transport model studies that normal 3-dimensional transport processes coupled with the known vertical N_2O mixing ratio gradient between the troposphere and stratosphere should provide a source of tropospheric variability. Therefore, given that variability was decreasing with improved methods of detection and measurement technique, that no tropospheric destruction mechanism was known, and that the stratospheric structure of N_2O would not be influenced by the nature of the tropospheric source as long as the net source balanced the stratospheric destruction, we designed a series of minimum variability N_2O experiments. Besides generating the stratospheric N_2O fields and transport information we sought, these experiments would help determine the need, if any, for destruction mechanisms other than those already known to exist in the stratosphere.

The first experiment, "Uniform Source" N_2O was a minimum tropospheric variability experiment with a uniform global surface layer source of 1.3×10^9 molecules $cm^{-2} sec^{-1}$ (about 15Mtons y^{-1}). Initially, the tropospheric source rate was set equal to the net estimated global destruction rate for a tropospheric surface mixing ratio of 0.295 ppm. The N_2O field was allowed to evolve and the source was adjusted to fit the calculated yearly mean destruction. The final mixing ratio and source strength may then be scaled to fit any desired surface value. The second experiment, "No-Source" N_2O was run from the equilibrium N_2O distribution generated by "Uniform Source" N_2O with the surface source removed. The remaining variability in the troposphere may then be directly attributed to the effect of transport from the stratosphere. The third experiment "Swamp Source" N_2O, was also started with the equilibrium distribution established by "Uniform Source" N_2O and had a constant surface land source (no ocean source) of 2.00×10^{10} molecules $cm^{-2} sec^{-1}$ only in those boxes in which the simulated annual rainfall exceeded an arbitrary minimum of 50 inches (127 cm). This led to a maximum source strength in the equatorial region, significant sources in the subtropics and northern mid-latitudes, no sources south of 50°S and north of 70°N, and a strong hemispheric asymmetry with the Northern Hemisphere source predominating. The global total source was identical to that for "Uniform Source" N_2O. This experiment then introduces the effect of discrete though small sources on tropospheric variability.

Model Results

The baseline experiment, "Uniform Source" N_2O, has run for seven model years, and, with the use of an efficient updating technique involving successive re-initializations, has reached both transport and chemical statistical equilibrium such that the yearly global destruction rate agrees to within 0.5% of the final equilibrium value. Seasonal zonal mean cross sections of N_2O mixing ratio and longitudinal relative standard deviation (V_λ) for the final model year are shown in Figure 1. A number of important features in the simulation, such as an almost uniform mixing ratio in the troposphere, a sharp poleward drop in mixing ratio along constant pressure surfaces in the stratosphere, and the penetration of tropospheric values into the lower stratosphere in the

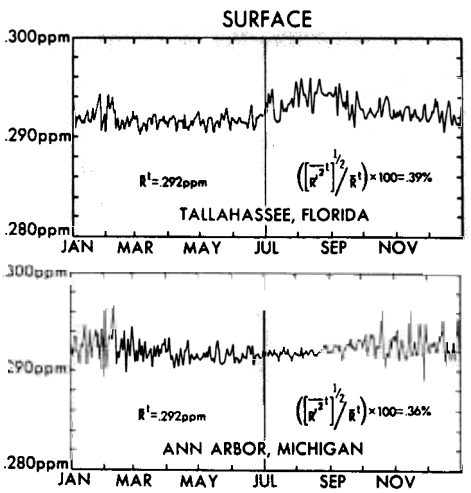

Fig. 2. One year time series of daily average N_2O mixing ratios from selected boxes. Yearly mean ($\overline{R^t}$) and temporal relative standard deviation (V_t) values are given as indicated. R' is the deviation of R from the yearly mean.

tropics, have been observed in the atmosphere (e.g., Schmeltekopf et al., 1977 and Pierotti and Rasmussen, 1977). More detailed model features, such as the minimums at 60°N and 50°S in the middle stratosphere during the winter and the 1-2% meridional variation in tropospheric mixing ratio with maximum values in the Southern Hemisphere and lowest values in the Northern mid- and high-latitudes, have not been confirmed in the real atmosphere.

It is interesting to note the strong correlation between longitudinal relative standard deviations and the vertical mixing ratio gradients shown in Figure 1. The region of minimum variability occurs in the tropospheric tropics and Southern Hemisphere where the mixing ratio is nearly uniform, while the stratosphere has much higher variability. Maximum variability occurs in the same region and at the same season that the mid-stratospheric minimums in mixing ratio are present. Longitudinal relative standard deviations in the surface layer range from 0.1-0.2% at the equator and subtropics to 0.3-0.5% at mid- and high-latitudes. They drop as low as 0.02% in the tropical mid-troposphere and rise in the stratosphere to values ranging from 2% at low latitudes to 9% at high latitudes. Overall, variability is higher in the model Northern Hemisphere.

One year time series of 24-hour averaged model N_2O mixing ratios for two surface boxes are shown in Figure 2. The dominating fluctuations are of synoptic scale (3-7 days). The temporal relative standard deviation exhibits the same general dependence on latitude and season seen in Figure 1. It ranges from 0.2% in the tropics to 0.8% in the high latitudes and compares well with 0.5% recently measured by Pierotti et al. (1978). The mid-latitude box containing Ann Arbor, Michigan reflects the effect of extratropical cyclones in the transport of stratospheric air from the lower stratosphere to the surface, thus increasing surface variability. The box containing Tallahassee, Florida shows a spring minimum which is caused by a transport of lower stratospheric N_2O poor air to that region. This transport feature is already observed in both the model and the real atmosphere as a spring maximum in surface ozone and radioactivity (Mahlman and Moxim, 1978). Temporal relative standard deviations in the stratosphere are much higher than in the troposphere with values running from 3% in the tropics to as high as 10-12% at mid- and high-latitudes.

The second N_2O experiment, "No-Source" N_2O, has been run for 4 model months from October to January. While not an experiment requiring source-sink equilibrations, it will be continued for at least one full year. Preliminary results show, as expected, no significant change in the stratosphere. The already low relative standard deviations have been halved in the middle troposphere and in the northern mid-latitude surface layer. The northern polar region as well as the tropics and Southern Hemisphere have had their relative standard deviations reduced by as much as 80%. A comparison between the "Uniform Source" N_2O and "No-Source" N_2O surface time series data is given in Figure 3. Preliminary model analysis indicates that the sporadic

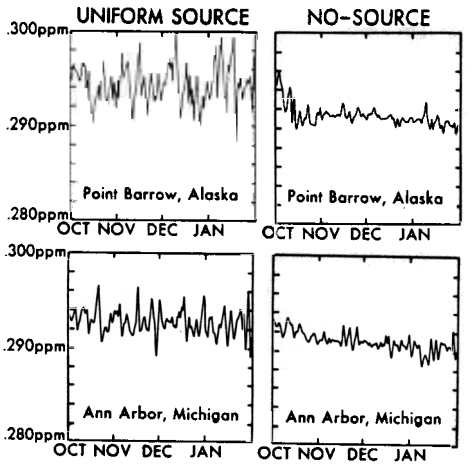

Fig. 3. Comparison between "Uniform Source " N_2O and "No-Source" N_2O time series of 24-hour averaged mixing ratios from selected surface boxes for the first 4 months of the "No-Source" experiment.

boundary layer accumulation of N_2O in stagnant anticyclones and its subsequent dispersal provides a major source of lower-troposphere variability. In the tropics, polar regions, and the summer Southern Hemisphere, it is the dominant source of variability, while in the winter northern mid-latitudes vertical transport from the lower stratosphere is still quite important.

The third experiment, "Swamp Source" N_2O, has just been started and run 6 months. While the preliminary results are not equilibrated for even the troposphere, a significant feature is the appearance of temporal relative standard deviations in the surface layer as high as 1% in tropical source regions. In non-source surface regions the values are as low as 0.1%. It appears that a much wider range of temporal and spatial variability in the lower troposphere is possible with discrete rather than uniform sources. Results from more detailed analyses of these three experiments will be presented in a later paper.

Summary

While the model N_2O mixing ratios are almost uniform in the troposphere, they do display a small south-to-north gradient. In general, the model stratosphere shows a sharp poleward drop along constant pressure surfaces, although mixing ratio minimums at 50°S and 60°N are observed in the winter middle stratosphere. The penetration of tropospheric mixing ratios into the tropical lower stratosphere is a prominent model feature.

The "Uniform Source" N_2O experiment generates surface longitudinal and temporal relative standard deviations in the range of 0.1-0.8% while some local surface values for the "Swamp Source" N_2O experiment exceed 1%. These results bracket the latest measurements of low N_2O relative standard deviations. While the model results do not disprove the existence of large troposphere sources and sinks, they do indicate that relative standard deviations of 0.5% or greater in the surface layer are possible with a small

tropospheric source of 15 Mton year^{-1} and a chemical lifetime of 150 years. It should be noted that, if the natural N_2O source is as small as 15 Mton year^{-1}, the main impact of man on stratospheric N_2O levels may not be through the indirect mechanism of increased nitrogen fixation, but through the direct anthropogenic production of N_2O (e.g., Weiss and Craig, 1976).

The Junge relationship between relative standard deviation and lifetime for a trace gas is not supported by the model data in the preceding paragraph and would appear to be inappropriate for such a long-lived trace gas.

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