

N95-11032

OBSERVED CHANGES IN THE VERTICAL PROFILE OF STRATOSPHERIC NITROUS OXIDE AT THULE, GREENLAND, FEBRUARY-MARCH, 1992

Louisa K. Emmons, John M. Reeves, Drew T. Shindell, and Robert L. de Zafta

Physics Department, State University of New York
Stony Brook, NY 11794 USA

Abstract

Using a ground-based mm-wave spectrometer, we have observed stratospheric N_2O over Thule, Greenland (76.3 N, 68.4 W) during late February and March, 1992. Vertical profiles of mixing ratio ranging from 16 to 50 km were recovered from molecular emission spectra. The profiles of early March show an abrupt increase in the lower-stratosphere N_2O mixing ratio similar to the spring-to-summer change associated with the break up of the Antarctic polar vortex. This increase is correlated with changes in potential vorticity, air temperature, and ozone mixing ratio.

Introduction

Nitrous oxide is an important tracer of transport, and in some cases an indicator of chemical processing,¹ in the stratosphere. Observations of N_2O distribution in the polar regions could further the understanding of polar atmospheric dynamics, yet it has not been aggressively studied. While aircraft and satellite measurements are vastly improving the body of polar N_2O data, complementary ground-based measurements can provide continuous, long-term monitoring of the vertical profile at virtually any latitude. As part of the Upper Atmosphere Research Satellite Correlative Measurements campaign and the European Arctic Stratospheric Ozone Experiment, we have measured the vertical profile of nitrous oxide at Thule, Greenland during February and March, 1992.

Briefly, the experimental method was the following: The emission intensity of the molecular rotation line at 276.328 GHz (1.08 mm) in N_2O was measured with a mm-wave heterodyne receiver/spectrometer whose mixing element is a superconducting (SIS) niobium tunnel junction. Details of the apparatus and data acquisition are given elsewhere.² Pressure-broadened emission spectra were measured in a bandwidth of 512 MHz at 1 MHz resolution, with emission intensity calibrated against black-body sources of known temperature. The pressure-broadened emission spectrum is not a simple Lorentzian

lineshape; rather, it is the sum of emission spectra from altitudes of differing ambient pressure, and pressure depends upon altitude. In the case of N_2O , the half-width at half maximum of molecules at 20 km is ~140 MHz, but falls to ~15 MHz by 35 km. The measured *composite* lineshape thus contains information on the vertical distribution of radiating molecules which is recoverable through numerical deconvolution. We extracted N_2O mixing ratios as functions of altitude with a modified Chahine-Twomey inversion algorithm,³ and used the N_2O pressure-broadening coefficient 2.38 MHz/mb reported by Lacombe et al.⁴

Observations and Discussion

The results of eleven separate N_2O observations spanning February 22 through March 25 are shown in Figure 1. Each curve, labeled at right by month and day in the format *md*, is the result of two hours of integration. Spectra are displayed as zenith-looking intensity, without atmospheric attenuation, expressed as an equivalent black-body radiation temperature. Each trace is offset 200 mK from the one below. The important feature is a marked increase in intensity, particularly near line center, following March 6 and persisting through March 25. The more pronounced peaking of these spectra indicate that the N_2O concentration increased in the middle and upper ranges of its distribution rather than everywhere uniformly in the stratosphere. This change is demonstrated in the vertical profiles of Figure 2. Lower and upper solid curves are typical mixing ratios recovered for the periods before and after March 7, respectively. Also plotted for comparison are a 1984 summer balloonsonde measurement from Antarctica⁵ (upper dashed line), and a vertical profile from McMurdo Station, Antarctica, obtained by mm-wave spectroscopy in the spring of 1986⁶ (lower dashed line). The data before March 7 yield a vertical profile much like that from the Antarctic spring, now established as indicating downward transport in a polar vortex region,⁶ while the late-March profile from Thule is similar to that of the Antarctic summer.

Our spectrometer bandwidth places a practical low-altitude limit on the recovery of vertical profiles, as highly pressure-broadened emission from lower in the atmosphere appears flatter and broader with decreasing altitude in the instrument's spectral window. Thus we have assumed a constant N_2O mixing ratio ~300 ppbv

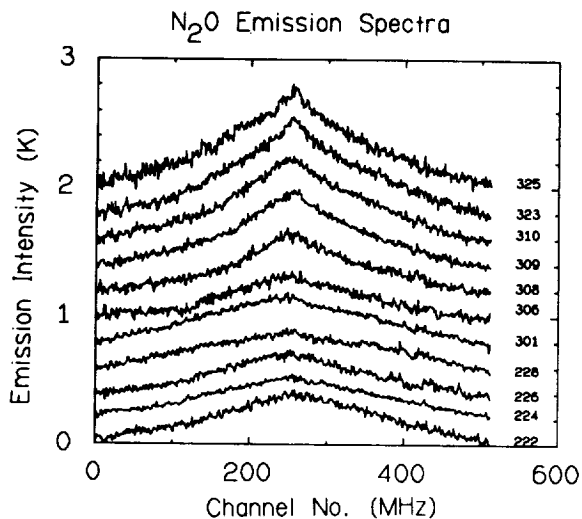


FIG. 1. N_2O emission spectra at 276 GHz, recorded on 11 separate days in February and March. Each trace, labeled at right by month and day (*mdd*), was averaged for two hours beginning about 23:00 local time. The vertical scale is relative emission intensity from the zenith direction expressed as an equivalent black-body radiation temperature, and 1 channel = 1 MHz on the horizontal axis. Curves are offset vertically by 200 mK.

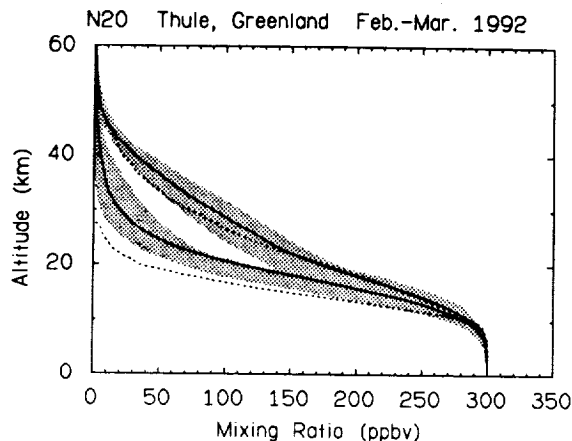


FIG. 2. N_2O vertical mixing ratio profiles. The lower (upper) solid curve is a profile recovered from emission intensity data before (after) March 7; shaded regions indicate the uncertainties in the distributions. The upper dashed curve is a balloonsonde measurement from the Antarctic summer, and the lower dashed curve is our own ground-based measurement from the Antarctic spring.

below 10 km, where the atmosphere is thoroughly mixed, for the deconvolutions shown. The accuracy of these profiles is limited by both noise and residual baseline deviations in the data; this uncertainty is indicated by shaded areas about each profile.

The increase in mixing ratio shown in Figure 2 occurs during a period around March 8 when stratospheric circulation over Thule switched from the periphery of the polar vortex (typically centered over Scandinavia and the Barents Sea) to flow across Canada from the Aleutian high. A transition can be clearly seen in plots of potential vorticity. In Figure 3, we show potential vorticity at three altitudes (potential temperature levels) for the days March 4 through March 11. The steepest potential vorticity gradient, marking the circulation edge of the polar vortex, is seen to have passed over Thule during March 7 to March 9. The increase in N_2O was also accompanied by changes in temperature and ozone distribution. Danish Meteorological Institute data for the same period show an increase from about 5 ppmv to 6.5 ppmv in the ozone profile from 20-35 km, and a 10 to 20 K increase in temperature from 10-30 km. While the ozone change was most pronounced between March 4 and March 11, the overall trend is one of fluctuation, so the increase is noted simply as further evidence of stratospheric change. Another indication of stratospheric change in early March is discussed in a companion paper on observed chlorine monoxide concentration over Thule.⁸

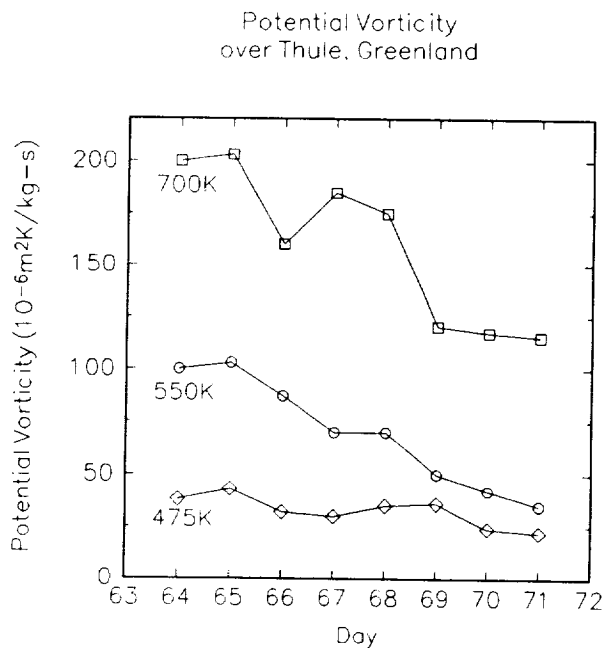


FIG. 3. Potential vorticity at three potential temperature levels over Thule, Greenland in early March. 700K approximately corresponds to 25 km, 550K to 22 km, and 475K to 19 km. The horizontal axis begins with March 3 = day 63. Data were provided by the Danish Meteorological Institute (DMI) through the Norwegian Institute for Air Research (NILU).

Conclusions

In addition to its use as a tracer of stratospheric transport and chemical processing, we believe N_2O concentration can be useful in defining the tropopause in the polar regions. There, the traditional tropopause marker of minimum temperature, or the sometimes-favored altitude at which ozone concentration reaches a minimum, may fail. Temperature profiles may not show a clear boundary, and O_3 may be too depleted at the altitude of interest. N_2O is uniformly distributed in the troposphere, but its mixing ratio drops with height above roughly 10 km due to photolysis and, at higher altitudes, reaction with 1D atomic oxygen. This destruction occurs slowly in the polar stratosphere, however, so the N_2O profile is determined mainly by air movement and hence is a good indicator of transport. The altitude at which the mixing ratio drops from its tropospheric value may therefore be the most readily-defined beginning of the polar stratosphere. Measurement of nitrous oxide concentration could provide a consistent definition of the tropopause which is largely independent of abnormal polar chemistry.

Acknowledgements

We would like to thank the Danish Meteorological Institute, particularly Bjorn Knudsen, and the Norwegian Institute for Air Research for the correlative O_3 and meteorological data used in this paper. We are grateful to the U.S. Air Force for permission to use the facilities at Thule Air Base and their cooperation during our stay. This work is supported by Grant NAGW-2182 through NASA's Upper Atmosphere Research Program and Grant NAG-1-1354 from the Upper Atmosphere Research Satellite Correlative Measurements Program.

References

1. S.E. Strahan, M. Loewenstein, J.R. Podolske, W. Starr, M.H. Proffitt, K.K. Kelly, and K.R. Chan, Correlation of N_2O and Ozone in the Southern Polar Vortex During the Airborne Antarctic Ozone Experiment, *J. Geophys. Res.* **94**, 413-423, 1980.
2. R.L. de Zafra, W.H. Mallison, M. Jaramillo, J.M. Reeves, L.K. Emmons, and D.T. Shindell, A New High-Sensitivity Superconducting Receiver for mm-Wave Remote-Sensing Spectroscopy of the Stratosphere, *these proceedings*.
3. S. Twomey, B. Herman, and R. Rabinoff, An Extension of the Chahine Method of Inverting the Radiative Transfer Equation, *J. Atmos. Sci.* **34**, 1085-1090, 1977.
4. N. Lacombe, A. Levy, and G. Guelachvili, Fourier Transform Measurement of Self-, N_2 -, and O_2 -Broadening of N_2O Lines: Temperature Dependence of Linewidths, *Appl. Optics* **23**, 425, 1984.
5. P.D. Goldan, W.C. Kuster, D.L. Albritton, and A.L. Schmeltekopf, Stratospheric $CFCl_3$, CF_2Cl_2 , and N_2O Height Profile Measurements at Several Latitudes, *J. Geophys. Res.* **85**, 413-423, 1980.
6. A. Parrish, R.L. de Zafra, M. Jaramillo, B. Connor, P.M. Solomon, and J.W. Barrett, Extremely Low N_2O Concentrations in the Springtime Stratosphere at McMurdo Station, Antarctica, *Nature* **332**, 53-55, 1988.
7. R.F. Weiss, The Temporal and Spatial Distribution of Tropospheric Nitrous Oxide, *J. Geophys. Res.* **86**, 7185-7195, 1981.
8. R.L. de Zafra, L.K. Emmons, J.M. Reeves, and D.T. Shindell, Measurements of the Vertical Profile, Diurnal Variation, and Secular Change of ClO in the Stratosphere over Thule, Greenland, February-March 1992, *these proceedings*.