

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

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**Abstract**

Using a ground-based mm-wave spectrometer, we have observed stratospheric N<sub>2</sub>O 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<sub>2</sub>O 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,<sup>1</sup> in the stratosphere. Observations of N<sub>2</sub>O 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<sub>2</sub>O 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<sub>2</sub>O 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.<sup>2</sup> 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<sub>2</sub>O, 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<sub>2</sub>O mixing ratios as functions of altitude with a modified Chahine-Twomey inversion algorithm,<sup>3</sup> and used the N<sub>2</sub>O pressure-broadening coefficient 2.38 MHz/mb reported by Lacombe et al.<sup>4</sup>

**Observations and Discussion**

The results of eleven separate N<sub>2</sub>O observations spanning February 22 through March 25 are shown in Figure 1. Each curve, labeled at right by month and day in the format *mdd*, 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<sub>2</sub>O 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 Antarctica<sup>5</sup> (upper dashed line), and a vertical profile from McMurdo Station, Antarctica, obtained by mm-wave spectroscopy in the spring of 1986<sup>6</sup> (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,<sup>6</sup> 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<sub>2</sub>O mixing ratio ~300 ppbv

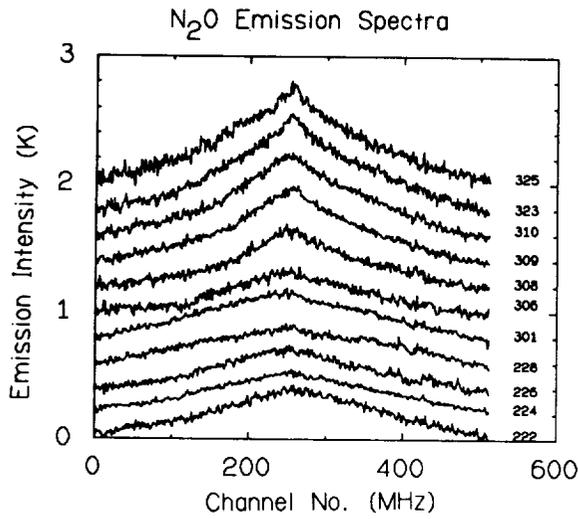


FIG. 1.  $N_2O$  emission spectra at 276 GHz, recorded on 11 separate days in February and March. Each trace, labeled at right by month and day (*mdd*), was averaged for two hours beginning about 23:00 local time. The vertical scale is relative emission intensity from the zenith direction expressed as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

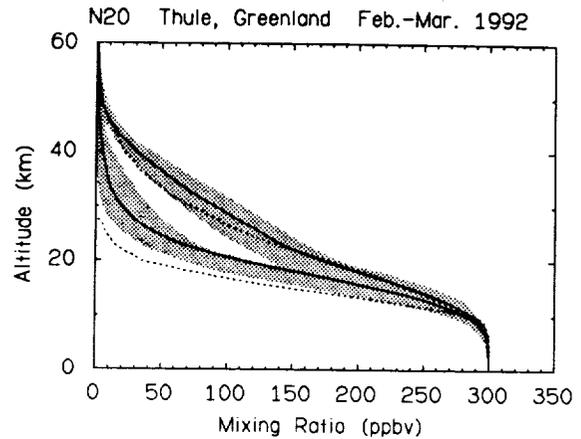


FIG. 2.  $N_2O$  vertical mixing ratio profiles. The lower (upper) solid curve is a profile recovered from emission intensity data before (after) March 7; shaded regions indicate the uncertainties in the distributions. The upper dashed curve is a balloonsonde measurement from the Antarctic summer, and 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.<sup>8</sup>

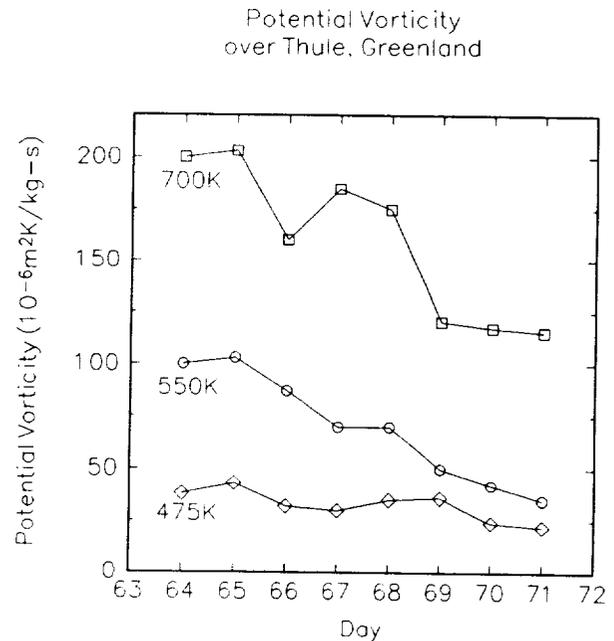


FIG. 3. Potential vorticity at three potential temperature levels over Thule, Greenland in early March. 700K approximately corresponds to 25 km, 550K to 22 km, and 475K to 19 km. The horizontal axis begins with March 3 = day 63. Data were provided by the Danish Meteorological Institute (DMI) through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

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