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20. We note that in our previous work, all absorption was treated in a purely linear fashion, as described in S. Solomon, A. L. Schmeltekopf, R. W. Sanders, *J. Geophys. Res.* **92**, 8329 (1987). The possible importance of nonlinear terms was also discussed in that work. The data analysis method used in this paper includes nonlinear terms and has decreased considerably the magnitude of unaccounted residual absorption features in the analysis. In view of this improvement in the data analysis, we have reanalyzed many of the lunar and solar spectra obtained in Antarctica (*9*). The improved analysis technique has not altered the results within their stated error bars. The details of the nonlinear inversion technique are described in A. L. Schmeltekopf *et al.*, unpublished results.
21. The indicated spectral range was measured with a diode array of 1016 independent diodes that were cooled to about −80°C. The image of the entrance slit on the array has a width of about 12 diodes, and an exit slit is applied by boxcar averaging over 12 diodes, providing an effective resolution of about 0.5 nm. The true electronic noise level in the system is about 0.01 to 0.03% for a typical count rate achievable in lunar spectra. Absorption features stronger than this should, in principle, be measurable with corresponding signal-to-noise.
22. Briefly, the spectral features of known components such as O_3, NO_2, Rayleigh scattering, and OClO were used in a coupled linear-nonlinear least-squares inversion that used all of the spectral features over the wavelength interval considered. The sum of the squares of residual absorption was minimized by the least-squares procedure and the resulting best-fit absorption of all components was retrieved. The remaining unaccounted for absorption is the residual spectrum. The standard error of the statistical fit to individual lunar spectra obtained at zenith angles greater than about 80° with this method is about 5 to 10%. We estimate that the total error on the measured vertical column abundance of OClO that we report is ±30%. The error bars in the figures on individual OClO measurements represent 1-σ standard errors of the spectral fit, and is a measure of the statistical precision of the spectral fit and not the total error.
23. All of the measurements we report were obtained with the direct light from the moon directed into the spectrometer with two front-surfaced mirrors, one on the roof of the trailer where the instrument was housed and the other set at a 45° angle to the entrance slit inside the building. The mirror on the roof was turned remotely to track the moon by means of stepper motors for each of the azimuth and zenith directions.
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Observations of Stratospheric NO_2 and O_3 at Thule, Greenland

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Scattered sunlight and direct light from the moon was used in two wavelength ranges to measure the total column abundances of stratospheric ozone (O_3) and nitrogen dioxide (NO_2) at Thule, Greenland (76.5°N), during the period from 29 January to 16 February 1988. The observed O_3 column varied between about 325 and 400 Dobson units, and the lower values were observed when the center of the Arctic polar vortex was closest to Thule. This gradient probably indicates that O_3 levels decrease due to dynamical processes near the center of the Arctic vortex and should be considered in attempts to derive trends in O_3 levels. The observed NO_2 levels were also lowest in the center of the Arctic vortex and were sometimes as low as 5×10^{14} molecules per square centimeter, which is even less than comparable values measured during Antarctic spring, suggesting that significant heterogeneous photochemistry takes place during the Arctic winter as it does in the Antarctic.

THE OBSERVATION OF A PRECIPITOUS decrease in Antarctic O_3 levels (*1*) has focused attention on the photochemistry of O_3 in both polar regions. Since the discovery of the Antarctic "ozone hole" in 1985, the status of Antarctica has changed from being the least explored portion of the earth's stratosphere to arguably the best explored. The current understanding of Antarctic photochemistry leading to the ozone hole is briefly summarized in a companion paper (*2*), wherein it was emphasized that similar phenomena may take place in the Arctic under those circumstances when the polar vortex is unusually cold and undisturbed (temperatures less than about −80°C). Such conditions are likely to lead to an unusual chemical system in which heterogeneous polar stratospheric cloud (PSC) chemistry plays an important role. As emphasized in (*2*), both extreme low temperatures and sunlight are required to drive the heterogeneous chemistry believed to be of central importance in establishing the Antarctic ozone hole.

Nitrogen dioxide plays a critical role in the photochemistry characterizing the Antarctic spring stratosphere. Greatly suppressed NO_2 levels are a necessary condition for halogen chemistry to effectively destroy O_3, since the partitioning between $ClONO_2$ and ClO is directly dependent on the NO_2 abundance. The column abundance of NO_2 is extremely low during the Antarctic spring (*3*, *4*) during the formation of the ozone hole. Very low NO_2 levels, highly elevated ClO and OClO levels, and substantial O_3 depletion result from heterogeneous chemistry occurring on PSCs in the sunlit Antarctic spring stratosphere [see references in (*2*)]. Thus measurements of stratospheric NO_2 along with observations of O_3 and chlorine free radicals are of great importance in considering the possibility of similar phenomena occurring in the Arctic.

Surprisingly little data are available on NO_2 in the Arctic lower stratosphere for winter and spring. Measurements by the Limb Infrared Monitor of the Stratosphere (LIMS) experiment provide the best seasonal and latitudinal coverage and reveal low abundances of NO_2 in the Arctic winter. However, the very low levels of NO_2 observed there approach the detection limit of the instrument for levels below about 10 mbar (*5*), and hence do not provide much information on the levels where PSC chemistry and subsequent O_3 depletion are likely to occur (from about 100 to 30 mbar, or about 15 to 25 km). Measurements by the Stratospheric Aerosols and Gases Experiments (SAGE) and Solar Mesosphere Explorer (SME) satellites can only be per-

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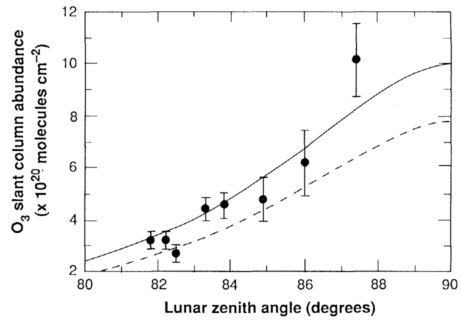

Fig. 1. Change in the O_3 slant column with increasing lunar zenith angle obtained in direct moon observations in the wavelength range from 400 to 453 nm on the night of 3 February 1988. Lines indicated the expected behavior for constant vertical column abundances of (– –) 300 and (——) 400 Dobson units. The background lunar zenith angle was ~70°.

formed in the fully sunlit atmosphere (*6*). Total O_3 observations by ground-based Dobson spectrophotometers and by backscatter ultraviolet techniques used by satellites are similarly largely limited to those seasons and latitudes when the sun is above the horizon.

Stratospheric sunrise begins when the sun is still below the horizon as seen from the surface (solar zenith angles of about 93° to 95° for altitudes near 10 to 25 km), leaving the troposphere in the darkness of the earth's shadow. Even this modest illumination is sufficient to drive important photochemical effects such as the photolysis of Cl_2 and HOCl (*2*). The scattered light technique involves detection of the light scattered by the sky, generally obtained by instruments pointing directly upward at the zenith sky (*7*). This method can be used to measure several stratospheric species when the sun is still several degrees below the horizon, allowing for high-latitude measurements to be made at a very early and particularly interesting point in the spring season in terms of possible heterogeneous photochemistry requiring both extreme cold temperatures and sunlight.

A few ground-based measurements that used scattered sunlight have been reported (*7*) for the Arctic. The current database on total column NO_2 is summarized in (*8*), wherein it is shown that the LIMS, SAGE, and SME instruments suggest total column abundances above: ~24 km near 70°N of about 1.5×10^{15} to 2.0×10^{15} cm^{-2}. Similar values were obtained on the average from the available ground-based measurements, based principally on observations obtained in Alaska (*7*). As noted in (*7*), stratospheric NO_2 above Alaska is subject to considerable variability due to the proximity of the Aleutian high, and the measurements reported were seldom very near to the center of the Arctic vortex. However, an important set of observations at relatively low latitudes (50°N) during winter, under conditions when Arctic air from the center of the cold vortex was rapidly transported to mid-latitudes, long ago pointed toward the possibility of significant removal of NO_2 through heterogeneous reactions in the Northern Hemisphere (*7*). Insights gained from the study of the Antarctic ozone hole have demonstrated the likely significance of those data, and defined the need for further measurements from the center of the vortex early in the spring season under the coldest possible conditions.

The observations we describe were obtained at Thule, Greenland (76.5°N), using visible and near-ultraviolet absorption spectroscopy. The choice of timing and location for the experiment, as well as the OClO observations acquired at night that used the moon as a light source, were described in (*2*). In this report, we discuss the observations of stratospheric O_3 and NO_2 obtained largely by using the scattered sunlight from the zenith sky as a light source. These data revealed the NO_2 and O_3 contents of the stratosphere near the center of the Arctic polar vortex while the sun was still slightly below the horizon and during a period when the temperatures near 50 to 30 mbar (near 20 km) were below −80°C, providing important information on the potential for Arctic heterogeneous chemistry and O_3 depletion.

The instrument, data analysis procedure, and sources of error have been discussed elsewhere (*3, 9*). Nearly all of the measurements we report were obtained in the spectral interval from 400 to 450 nm (*3*) and were acquired by using scattered daylight. The measurements made by using scattered sunlight must be interpreted with calculated "air mass factors" that use scattering theory to account for the path of incoming light (*7, 9*). Measurements of total O_3 by using direct light from the moon in the spectral region from 330 to 345 nm are also presented for the night of 31 January 1988 for comparison [see (*9*)]. The use of this other spectral interval provides an important check for the measurements in which the weaker O_3 absorption features near 400 to 450 nm were used. Furthermore, measurements in which the direct light from the moon or sun was used provided an important check on the scattered light measurements, since the accuracy of the latter type of measurement is dependent on the accuracy of the scattering model used to derive air mass factors, whereas the air mass factors characterizing direct light measurements are obtained directly from simple geometrical considerations.

Zenith sky spectra were obtained daily beginning at Thule on 29 January 1988 and ending on 16 February 1988. During the first week of these measurements, the sun was well below the horizon even at noon, which is important for two reasons: (i) Rather long integration times were required to obtain adequate count rates. The integration times were sometimes as great as 6500 s and never less than 900 s. (ii) The air mass factors appropriate to solar zenith angles greater than 93° are subject to greater uncertainty than those at smaller angles because of the importance of multiple scattering at large angles (*7, 9*). The solar zenith angle at noon became less than 93° on 5 February. The scattered light measurements obtained prior to this date are thus subject to larger uncertainties than those obtained later. Solar zenith angles as small as 89.3° were obtained in mid-February.

The analysis of all of the data was carried out with the linear least-squares method used in our earlier studies (*3, 9*) and did not include the coupled linear-nonlinear approach as in the companion paper (*2*). The scattered light measurements were all reduced by using a background spectrum derived for conditions outside the earth's atmosphere by extrapolation (*2, 9*) and hence provide a measure of the total slant column of absorber. This analysis technique yielded vertical NO_2 and O_3 abundances in Antarctica that were in good general agreement with simultaneous observations by other methods (*3, 4, 9*). In measurements made at McMurdo Station during 1986, the stan-

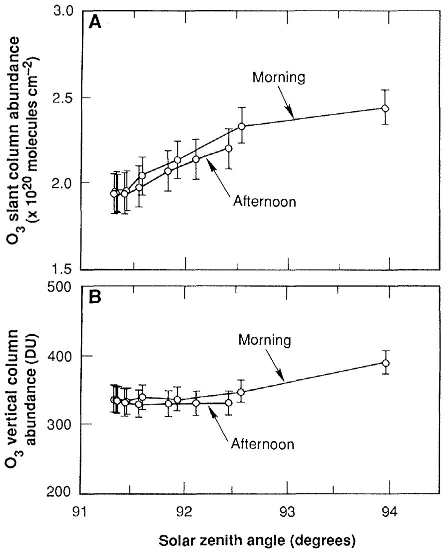

Fig. 2. (**A**) Observation of the O_3 slant column abundance versus solar zenith angle obtained with scattered light from the zenith sky on 9 February 1988. (**B**) Observations of the vertical column abundance of O_3 derived from the slant column measurements in (A).

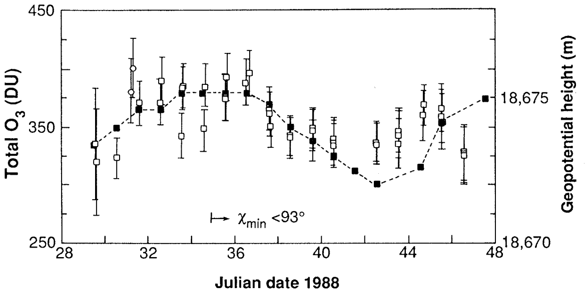

Fig. 3. Representative total O_3 column measurements obtained daily at Thule from 29 January to 16 February 1988: (□), visible spectroscopy, ~425 to 450 nm, zenith sky; (○), near-ultraviolet spectroscopy, ~330 to 340 nm, direct moon. The geopotential height at the 70-mbar level (about 18 km) taken from NMC analyses (■) is plotted for comparison.

dard deviation of the difference between such measurements of O_3 and those made with high-resolution infrared spectroscopy was 11.0%, whereas the average difference between the two methods was only 3.2% (*9*). The precision of the spectral fit to the scattered light measurements of total O_3 in the region from 400 to 450 nm is about 10 to 15%, whereas the absolute accuracy is estimated to be ±25%. The precision and accuracy of the lunar O_3 measurements made in the visible part of the spectrum is not as good as the scattered light measurements, since the air mass factors are much smaller and hence the total absorption much weaker. The precision of the NO_2 measurements is of the order of 5%, and the absolute accuracy is estimated at ±25% (*3, 9*). In the figures we present, error bars indicate the precision of the spectral fit and not the total errors given above.

Measurements of the slant column of O_3 observed on the night of 4 February 1988 at Thule with the direct moon as a light source are shown in Fig. 1. The data were reduced by using a lunar spectrum obtained at a lunar zenith angle of about 65° as the background spectrum for measurements made at larger angles, as is described in (*2*). The dashed and solid lines show the expected response by assuming constant total vertical column abundances of 300 and 400 Dobson units (DU; 1 Dobson unit = 10^{-3} atm cm), respectively. The data show that the total O_3 column abundance was ~350 DU on this particular night. Similar values were obtained, with the use of the spectral interval from 330 to 345 nm on the night of 31 January 1988 (see below), providing important verification with a different set of spectral features (*9*).

Measurements of the O_3 slant column versus solar zenith angle obtained on 9 February 1988 that used scattering from the zenith sky as the light source are shown in Fig. 2A. The observed values increased systematically with increasing angle due to the increase in air mass (*3, 7, 9*) as the sun sets, and the shape of the growth as a function of angle provides information on the altitude where the bulk of the absorber resides. The corresponding vertical column abundances assuming that the O_3 can be approximately described as a Gaussian layer with 7-km half-width centered at 15 km (*3, 9*) are shown in Fig. 2B. These data suggest that the O_3 column is centered near 15 km, and that the total vertical column abundance is about 350 DU, in broad agreement with the direct-light measurements shown in Fig. 1.

The time series of total O_3 measurements during the observing period is shown in Fig. 3. For clarity, only a limited number of data points are shown for any particular date rather than all of the points. As shown in Fig. 2B, excellent agreement was obtained among O_3 measurements made at various solar zenith angles on any given day. A dashed line indicates the geopotential height of the 70-mbar level as derived from the National Meteorological Center (NMC) analyses. A general correspondence between geopotential height and total O_3 was observed, with lower total O_3 being found when the center of the vortex moved closer to Thule, as indicated by low geopotential height. This effect probably indicates the normal gradient of total O_3 and should be considered in any attempt to detect Arctic O_3 depletion by examining O_3 trends.

Measurements of stratospheric NO_2 can also be carried out with both scattered and direct light. The first night of our moon observations at Thule revealed the presence of tropospheric NO_2 pollution. Unlike the O_3 observations shown in Fig. 2, moonlit NO_2 observations did not display a systematic pattern as a function of lunar zenith angle. Large variability was observed, with corresponding vertical column amounts ranging from a few times 10^{14} cm^{-2} to as much as 2.0×10^{16} cm^{-2}. The latter values surely resulted from tropospheric pollution, due perhaps both to local pollution from the station power plant and to the general phenomenon known as "Arctic haze." Pollution emanating from populated regions spreads throughout the Arctic in winter [see, for example, (*10*)] and renders the moon measurements useless for stratospheric NO_2 observations.

However, the weighting for NO_2 in the stratosphere as compared with the troposphere is far better for scattered light measurements than it is for direct light measurements because of the differences in optical paths (*7, 9*) between the two for scattered light at large solar zenith angle. Thus the scattered light measurements are more useful for measurements of NO_2 in highly polluted environments than are direct light measurements. After 7 February, the sky was bright enough for sufficiently extended periods that as many as 7 to 20 spectra could be collected in a single day. These spectra can be readily examined for evidence of the variability likely to be induced by tropospheric pollution. As an example the observations obtained on 15 February assuming a Gaussian layer centered at 25 km are shown in Fig. 4. There are several periods when large, variable NO_2 abundances were obtained (presumably because of tropospheric pollution), but there is also an underlying consistent value that probably represents the stratospheric component. At worst, these measurements provide a realistic upper limit to the NO_2 present in the stratosphere.

The time series of available NO_2 measurements is shown in Fig. 5. The data were visually inspected as shown in Fig. 4 to determine which measurements were likely to be stratospheric for all days after 7 February (day 38). Prior to this date, the points indicated in the Fig. 5 should be considered upper limits to the stratospheric abundances. Representative points are plotted as in Fig. 3. Noon values were used where

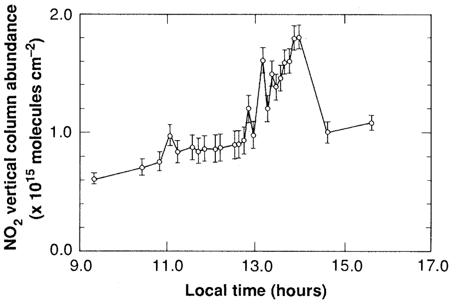

Fig. 4. Total NO_2 column abundance observed at Thule by using scattered sunlight on 15 February 1988. Short-lived variations are likely due to tropospheric pollution.

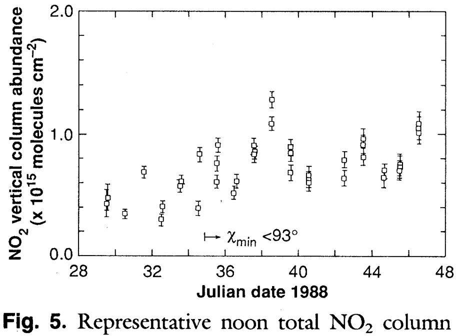

Fig. 5. Representative noon total NO_2 column abundances measured daily at Thule; visible spectroscopy, ~425 to 450 nm.

unpolluted. Like the O_3 measurements shown in Fig. 3, the NO_2 measurements also reveal a decrease near day 40 to 42 when the center of the geopotential low was nearest to Thule, as indicated in Fig. 3, suggesting that the NO_2 abundances near the center of Arctic vortex are less than those outside. This result supports the assertion that these data are dominated by stratospheric NO_2 rather than tropospheric pollution.

The observed values and upper limits for NO_2 shown in Figs. 4 and 5 are extremely low. They are as low and lower than the Antarctic measurements during August at McMurdo Station (*3, 4, 9*). For comparison, two-dimensional model calculations (*11*) predict a total NO_2 column of 1.5×10^{15} cm^{-2} at the end of January and 2.0×10^{15} cm^{-2} at the end of February for 75°N. Thus these observations suggest that a significant depletion of stratospheric NO_2 had occurred at Thule during the winter of 1988. This result is not particularly surprising in view of the extreme low temperatures over Thule during this period and in view of theoretical and observational studies of HNO_3 suggesting that nitrogen radical species are converted to HNO_3 during the polar winter (*12*), probably through heterogeneous chemistry. Further interpretation of such observations requires detailed consideration of the past history of the air parcels over Thule so that the formation and destruction of N_2O_5 can be evaluated, as well as the frequency and chemistry of PSCs.

Few in situ measurements of nitrogen radical species in winter at high latitudes are available. Balloon-borne measurements reveal NO and NO_2 mixing ratios of about 60 to 100 parts per trillion by volume (pptv) and 100 to 150 pptv, respectively, from 15 to 22 km (*13*) in western Canada under relatively warm winter conditions (that is, temperatures of $-50°$ to $-60°C$ from 15 to 30 km). Thus these measurements were conducted under much warmer conditions and much farther from the center of the Arctic vortex than the observations reported here. Simultaneous ground-based measurements of the total NO_2 column during the period of those observations range from about 0.7×10^{15} to 2.0×10^{15} cm^{-2} (*13*). The latter values were typical of afternoon twilights and are significantly greater than the evening twilight values measured here. The low total column NO_2 measurements at Thule thus suggest that the abundances of lower stratospheric NO_x were likely well below 60 pptv, which is consistent with the assertion that the observation of nighttime OClO abundances of about 5×10^{13} cm^{-2} as presented in (*2*) imply NO_2 mixing ratios of only a few parts per trillion by volume in the lower stratosphere. These considerations, together with the observation that NO_2 levels over Thule were even lower than our measurements performed at McMurdo Station, Antarctica, suggest that heterogeneous chemistry plays an important role in determining the abundance of nitrogen and chlorine species during cold Arctic winters.

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In Situ Northern Mid-Latitude Observations of ClO, O_3, and BrO in the Wintertime Lower Stratosphere

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In order to test photochemical theories linking chlorofluorocarbon derivatives to ozone (O_3) depletion at high latitudes in the springtime, several related atmospheric species, including O_3, chlorine monoxide (ClO), and bromine monoxide (BrO) were measured in the lower stratosphere with instruments mounted on the NASA ER-2 aircraft on 13 February 1988. The flight path from Moffett Field, California (37°N, 121°W), to Great Slave Lake, Canada (61°N, 115°W), extended to the center of the polar jet associated with but outside of the Arctic vortex, in which the abundance of O_3 was twice its mid-latitude value, whereas BrO levels were 5 parts per trillion by volume (pptv) between 18 and 21 kilometers, and 2.4 pptv below that altitude. The ClO mixing ratio was as much as 65 pptv at 60°N latitude at an altitude of 20 kilometers, and was enhanced over mid-latitude values by a factor of 3 to 5 at altitudes above 18 kilometers and by as much as a factor of 40 at altitudes below 17 kilometers. Levels of ClO and O_3 were highly correlated on all measured distance scales, and both showed an abrupt change in character at 54°N latitude. The enhancement of ClO abundance north of 54°N was most likely caused by low nitrogen dioxide levels in the flight path.

TRACE CONSTITUENTS IN THE POLAR regions of the stratosphere have until recently received little attention, partly because the photochemical processes there were thought to be negligible compared with the better illuminated mid-latitudes, and partly because observing platforms for trace species have only rarely sampled there. The discovery of the rapid O_3 depletion over Antarctica in the austral spring (*1*) has focused attention on the interaction between photochemistry and transport of trace species at high southern latitudes. Initially the discussion about the mechanism for O_3 depletion centered on either a chemical (actual O_3 loss) or dynamical (O_3 redistribution) cause (*2*). However, results from ground-based observing missions to Antarctica, NOZE I (*3*) in 1986 and NOZE II (*4*) in 1987, and from the Airborne Antarctic Ozone Experiment

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