Lower atmosphere studies

The National Ozone Expedition, 1986

SUSAN SOLOMON

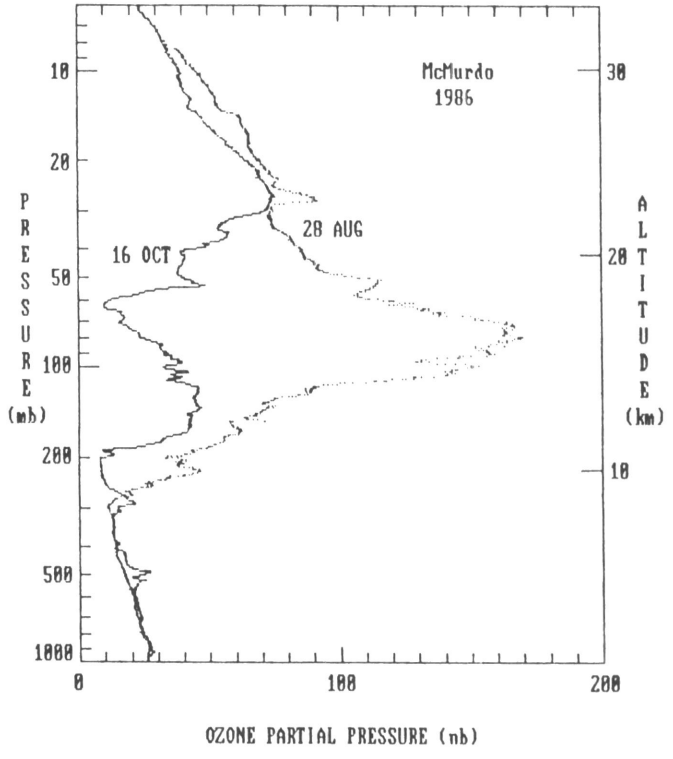
National Oceanic and Atmospheric Administration
Aeronomy Laboratory
Boulder, Colorado 80303

A remarkable change in the antarctic ozone layer was first established in a study by the British Antarctic Survey (Farman, Gardiner, and Shanklin 1985). Their observations at Halley Bay demonstrated that the total ozone abundance had decreased by about 50 percent during the antarctic spring seasons of the past decade. This constitutes a variation in total ozone that far exceeds natural variability observed previously anywhere in the world, and has prompted a great deal of concern regarding the cause and ramifications of such an extreme environmental perturbation. Both dynamical (Tung et al. 1986; Mahlman and Fels 1986) and chemical (Callis and Natarajan 1986; McElroy et al. 1986; Solomon et al. 1986) theories were advanced as possible explanations for this phenomenon. All the theories agree that the unique appearance of this phenomenon in the Antarctic is related to the extreme coldness of the antarctic atmosphere during winter and spring, perhaps through processes involving polar stratospheric clouds, sometimes visible in the twilight sky in the Antarctic.

Eighteen scientists from four separate institutions came to McMurdo Station during the period from August to November, 1986, to carry out an intensive stratospheric measurement program aimed at obtaining further data on the antarctic ozone "hole." The results from the composite of experiments strongly suggest that chemistry (specifically, the chemistry of anthropogenically produced halocarbon species) probably plays an important role in the development of the antarctic ozone hole. If the antarctic ozone hole is due to mankind's use of chlorofluorocarbons, then it represents the first time that the environment has been shown to be sensitive to man's activities on a global scale.

Balloon observations by the group from the University of Wyoming established that the vertical domain of the total ozone hole is quite restricted, extending only from about 10 to 22 kilometers as shown in the figure (from Hofmann et al. 1987). Both these observations and the observations of ozone by visible spectroscopy by the National Oceanic and Atmospheric Administration's Aeronomy Laboratory showed that the ozone hole develops rapidly during early September. The group from the University of Wyoming also performed several simultaneous balloon measurements of the vertical distributions of particulate matter and ozone in the stratosphere. They concluded that those data implied that dynamical transport processes are probably not the cause of the ozone hole (Hofmann et al. 1988). Observations of nitrous oxide by the microwave emission instrument of the group from the State University of New York at Stony Brook (Parrish et al. in press) also suggest that dynamical processes are not likely to be the direct cause of the depletion of antarctic ozone.

Observations of chlorine-containing molecules at McMurdo displayed extremely unusual characteristics. Chlorine monoxide plays a central role in all halocarbon theories of the hole. This species was observed by the Stony Brook group and found to be present in abundances about 50–100 times greater than "standard" theoretical models near the 20-kilometer level (de Zafra et al. 1987; P. Solomon et al. 1987). Chlorine dioxide is a closely related species. Observations of chlorine dioxide by the National Oceanic and Atmospheric Administration group also displayed abundances that were about 50 times greater than standard models (S. Solomon et al. 1987), in excellent agreement with the observations by the Stony Brook group. The two observations taken together provide strong evidence that the chlorine chemistry of the antarctic lower stratosphere is highly



Ozone partical pressure profiles at the start of ozone hole formation and at the height of the depression over McMurdo Station in 1986. Temperatures in the 14–18-kilometer region were −80°C or lower in both cases, indicating that McMurdo was well inside the polar vortex at these times. From Hofmann et al. (1987). ("mb" denotes "millibars." "nb" denotes "nanobars." "km" denotes "kilometers.")

disturbed, and probably plays an important role in the development of the antarctic ozone hole.

Several outstanding questions remain in the study of the antarctic ozone phenomenon, and the issue is far from being fully resolved at this writing. Further work on the role and chemistry of polar stratospheric clouds is badly needed, as are earlier measurements during the antarctic winter so that the temporal development of the hole can be studied in detail.

References

Callis, L.B., and M. Natarajan. 1986. The Antarctic ozone minimum: Relationship to odd nitrogen, odd chlorine, the final warming, and the 11 year solar cycle. *Journal of Geophysical Research*, 91, 10771–10798.

de Zafra, R.L., M. Jaramillo, A. Parrish, P. Solomon, B. Connor, and J. Barnett. 1987. Observation of abnormally high concentrations of chlorine monoxide at low altitudes in the Antarctic spring stratosphere. *Nature*, 328–408.

Farman, J.C., B.G. Gardiner, and J.D. Shanklin. 1985. Large losses of total ozone in Antarctica reveal seasonal ClO_x/NO_x interaction. *Nature*, 315, 207–210.

Hofmann, D.J., J.W. Harder, S.R. Rolf, and J.M. Rosen. 1987. Balloon-borne observations of the temporal development and vertical structure of the Antarctic ozone hole in 1986. *Nature*, 326, 59–62.

Hofmann, D.J., J.M. Rosen, and J.W. Harder. 1988. Aerosol measurements in the winter/spring Antarctic stratosphere: I. Correlative measurements with ozone. *Journal of Geophysical Research*, 93, 665.

Mahlman, J.D., and S.B. Fels. 1986. Antarctic ozone decreases: A dynamical cause. *Geophysical Research Letters*, 13, 1316–1319.

McElroy, M.B., R.J. Salawitch, S.C. Wofsy, and J.A. Logan. 1986. Antarctic ozone: Reductions due to synergistic interactions of chlorine and bromine. *Nature*, 321, 759–762.

Parrish, A., R.L. DeZafra, M. Jaramillo, B. Connor, P.M. Solomon, and J. Barrett. In press. Observations of extremely low N_2O concentrations in the springtime stratosphere at McMurdo station, Antarctica. *Nature*.

Solomon, P., B. Connor, R. de Zafra, A. Parrish, J. Barrett, and M. Jaramillo. 1987. Observations of abnormally high concentrations of chlorine monoxide at low altitudes in the Antarctic spring stratosphere II. Secular variation. *Nature*, 328, 411.

Solomon, S., R.R. Garcia. F.S. Rowland, and D.J. Wuebbles. 1986. On the depletion of Antarctic ozone. *Nature*, 321, 755–758.

Solomon, S., G.H. Mount, R.W. Sanders, and A.L. Schmeltekopf. 1987. Visible spectroscopy at McMurdo Station, Antarctica 2. Observations of OCIO. *Journal of Geophysical Research*, 92, 8329.

Tung, K.K., M.K.W. Ko, J.M. Rodriquez, and N.D. Sze. 1986. Are Antarctic ozone variations a manifestation of dynamics of chemistry? *Nature*, 333, 811–814.

Observation of stratospheric trace gases related to ozone depletion in the antarctic spring

R.L. DE ZAFRA, A. PARRISH*, P. SOLOMON, J.W. BARRETT, B. CONNOR**, *and* M. JARAMILLO

State University of New York at Stony Brook
Stony Brook, New York 11790

During the first National Ozone Expedition (NOZE I), which ran from 21 August to early November 1986 at McMurdo Station, we made frequent measurements of chlorine monoxide (ClO), ozone (O_3), nitrous oxide (N_2O), and occasional measurements of hydrogen cyanide. Observations were made with a ground-based millimeters wave spectrometer capable of detecting and measuring the pressure broadened rotational emission lines of these molecules in the 260–280 gigahertz frequency range (Parrish et al. 1988). The spectral bandpass and resolution of the instrument is sufficient to recover altitude distributions over a range of approximately 20–55 kilometers and to detect emission from as low as approximately 13–15 kilometers.

NOZE I was organized to give the first systematic look at a number of factors potentially involved with the seasonally occurring antarctic ozone hole (Farman, Gardiner, and Shanklin 1985). Of primary concern were several stratospheric trace gases known to be involved in regulating the formation and destruction of ozone in the normal stratosphere. Chemical theories attempting to explain the formation of the ozone hole have, as a common denominator, the prediction of far more ClO at low altitudes than is normally found there. The details of the chemical interactions involved vary from theory to theory (McElroy, et al. 1986; S. Solomon, et al. 1986; Rodriquez, Ko, and Sze 1986; Crutzen and Arnold 1986; Molina and Molina 1987), but all predict a large excess of ClO in the same altitude range and geographical region where ozone is undergoing significant depletion.

Measurements of chlorine monoxide. The observation of excess ClO at low altitudes in Antarctica would be a clear sign of a primary role played by chlorine chemistry in the formation of the ozone hole and would at least diminish the importance of other theories relying on dynamics (e.g., Tung 1986; Mahlman and Fels 1986), sunspot cycles (Callis and Natarajan 1986), etc. (The ultimate source of most stratospheric chlorine is now the photolysis of chlorofluorocarbons produced for a variety of commercial purposes.) Our measurements at McMurdo have revealed just such an excess of low-altitude ClO, amounting to approximately 100 times more than normally found at an altitude of 20 kilometers. The altitude range within which the greatest depletion of ozone takes place—14–18 kilometers—is too low to allow details of the vertical distribution of detected ClO to be recovered with the instrument used during NOZE I, but its presence is unmistakable in the data collected. ClO in the normal stratosphere has a strong diurnal variation, with greatest amounts in the daytime, and a strong decrease at night (P.M. Solomon et al. 1984, and references therein). A strong diurnal variation is also predicted from the chemistry postulated for the low-altitude component involved in the ozone hole, and this is clearly seen in our data (figure 1). In addition to the diurnal variation, it would be expected that the amount of low-altitude

* Also with Millitech Corporation, South Deerfield, Massachusetts.
** Present address: *Science and Technology Corporation, Hampton, Virginia.*