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PREPRINT

INFLUENCE OF A SOLAR PROTON EVENT ON STRATOSPHERIC OZONE

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Atmospheric ozone is believed to be destroyed in the stratosphere and mesosphere by catalytic agents in addition to well-known reactions with oxygen allotropes. The relative contributions to destruction by the catalysts are predicted to be altitude dependent with odd-hydrogen components (HO_x) dominant in the mesosphere and troposphere and odd nitrogen (NO_x) and halogens dominant in the stratosphere.

A measurement of mesospheric ozone changes associated with the solar proton event of November 2, 1969 was reported by Weeks, et al.[1] The first of two rockets carrying UV ozone photometers was fired into the initial phase of the event at Fort Churchill, Manitoba; and the second rocket was launched two days later under quieter conditions. The ozone concentration was lower during the event by a factor of 2 at 54 km and by a factor of 4 at 67 km, compared with the later flight. The changes and the rapid recovery are consistent with odd-hydrogen chemistry in the mesosphere.

Experiments which could confirm the catalytic reaction cycles in the stratosphere have been difficult to perform and analyze. The effects of gradual introduction of contaminants can be determined only if the natural variations of ozone are known over appropriate time periods. Transient injections with well defined spatial and temporal signatures, however, remove the ambiguity with the natural variability. Attempts to observe the local effects of nitric oxide from nuclear weapons tests were reported by Miller et al.[2] and Christie.[3]

High energy solar proton events are capable of impulsively depositing large amounts of energy into the polar cap regions at all latitudes above 55°. It has been pointed out by Crutzen et al.[4] that nitric oxide can be produced in the stratosphere during intense solar proton events and that such large scale production of nitric oxide[5] should be followed by reductions in ozone concentrations in the affected regions.

The first observations of an ozone decrease in the stratosphere associated with a solar proton event are reported here. That event on August 4, 1972, was the most intense of any recorded in the last 25 years.

The ozone measurements were made with the Backscattered Ultraviolet (BUV) experiment[6] on the Nimbus 4 satellite which was launched into a sun synchronous circular orbit with an inclination of 100° in April 1970. This experiment provides vertical ozone profiles every 200 km along the orbital track from the ozone maximum (25 mb) up to 0.4 mb by mathematical inversion of the earth albedo at selected wavelengths[7] and, in addition, measurements of total ozone[8].

The daily zonally-averaged ozone amounts above the 4 mb pressure surface are shown in Figure 1 for three geodetic latitude bands during July (Day No. 183-213) and August (Day No. 214-244), 1972. The highest latitude band (75-89° N) is above the geomagnetic cutoff latitude for the event; the other bands (55-65° N and 5° S-5° N) are magnetically protected, respectively.

The event, which occurred late on Day 217 (August 4), produced an abrupt ozone decrease in the 75-80°N zone of about 0.002 atm-cm above 4 mb, which apparently persisted throughout the month of August. The 55-65° N zone suffered a decrease on Days 219 and 220 but recovered on Day 221. Thereafter, a more gradual decrease was observed. The equatorial zone also showed gradual decreases after Day 218. The latter changes are not uniquely distinguishable from seasonal variations. No significant change was observed in the global ozone fields which could be attributed to major optical solar flares which occurred during the August 1972 period of intense solar activity.

The observed change in total ozone following the event was -0.003 atm-cm for the 75-80° N zone. This corresponds to a 1.3% reduction in the 0.305 atm-cm zonal average, but is well within the 0.019 atm-cm standard deviation. Thus the event was not observed from Dobson stations[9].

A better estimate of the total depletion in the 75-80° N zone is the change above the 10 mb surface. There a decrease of 0.004 atm-cm may be compared with the standard deviation of 0.001 atm-cm.

The production and deposition of NO at high geomagnetic latitudes during this event has been estimated using solar proton flux and energy data obtained from satellites.[10] The effect of this pulse of NO input on stratospheric ozone concentrations was calculated using a time-dependent, two-dimensional model which simulates the most important photochemical reactions and zonally-averaged transport in the stratosphere.[11] A vertical profile of the calculated reduction in

ozone concentrations resulting from NO catalysis for three weeks after the solar proton event of August 4, 1972, at 80° N is shown in Figure 2.[12]

The observed reductions in the ozone partial pressure as a function of air pressure for the 75-80° N zone are shown on the same figure. The data are derived from the average for the seven days preceding the event (Day 211-217) compared with seven day averages centered on eight and nineteen days after the event.[13] The observed and predicted ozone changes are in reasonable agreement with respect to the pressure level of maximum change (2 mb). The calculated decrease of 16% is about 30% less than the observed reductions.[14] On the other hand, the model calculations also showed reductions of ozone concentration down to the 30-40° latitude circle (not shown here), which could not be detected from the satellite. It is, therefore, clear that horizontal mixing in the 35-45 km region was overestimated in the model. Another interesting observational feature shown in Figure 2, which requires explanation, is the apparent relative increase in ozone reduction in the stratosphere from day 8 to day 19 after the event. These are typical examples of how continued analysis of the observed ozone distributions can supply essential new information on badly known mixing and on photochemical processes in the stratosphere.

If the current understanding of stratospheric transport and photochemistry is correct, in the summer season above 10 mb the ozone concentrations should be determined mainly by the effect of the photochemical reactions, especially those of the oxides of nitrogen NO and NO_2 for levels below 1 mb. The production rates of NO below the 4 mb surface are somewhat uncertain due to inadequate knowledge of the proton fluxes at high energies. However, almost all the NO produced by the solar proton event was injected into the stratosphere above 10 mb. The uncertainties in the deposition profile and in atmospheric transport are likely to be responsible for the differences in the 10 mb region.

The effect of increased production of HO_x in the mesosphere was not considered in the model[15]. As can be noticed from the observational results in

Figures 1 and 2, the recovery is slow below 1 mb. This finding is compatible with the expected long chemical removal time of NO_x in the stratosphere and with relatively slow transport of NO_x out of the production region.

The good agreement between observed and calculated ozone reductions at high latitudes and altitudes in the Northern Hemisphere following the August 1972 solar proton event constitutes, therefore, a first and convincing validation of the current photochemical theory of stratospheric ozone above 30 km.

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11. Included in the model are the most important photochemical reactions which determine the concentrations of O, O_3, N_2O, N, NO, NO_2, NO_3, N_2O_5, HNO_3, CH_4, CO, H_2, H, OH, HO_2, H_2O_2. Rate coefficients for reactions were those recommended by D. Garvin and R. F. Hampson, Eds., Chemical Kinetics Data Survey (Report NBSIR 74-430, National Bureau of Standards, Washington, D. C. 1974). Temperature and density data were adopted from CIRA 1965 (Cospar Working Group 4, Cospar International Reference Atmosphere, North-Holland, Amsterdam, 1965) and from A. H. Oort and E. M. Rasmusson (Atmospheric Circulation Statistics, NOAA Professional Paper, U. S. Department of Commerce, Rockville, Maryland, September 1971). The model extends from ground level to approximately 55 km and from South Pole to North Pole. Production of NO above 55 km could not be taken into account in the present model, i.e., downward flux of NO through the upper boundary was neglected. This neglect is not expected to affect critically calculated ozone concentrations during the first three weeks

following the solar proton event, as NO and NO_2 do not affect ozone concentrations significantly above about 45 km. Zonally averaged concentrations of chemical compounds were calculated adopting mean meridional circulation data (J. F. Louis, Ph. D. thesis, University of Colorado, 1974) and empirically determined time dependent eddy diffusion coefficients (P. J. Crutzen, presented at 4th CIAP Conference, Cambridge, Massachusetts, February 1974, in press).

12. P. J. Crutzen, XVI IUGG General Assy., Grenoble, France, August 25-September 6, 1975.

13. Days 222-225 and 227-229 are used for the +8 day mean. Day 226 was excluded because of a small sample size. Days 233-239 are used for the +19 day average. Theoretical calculations were made for Day 244 (September 1, 1972).

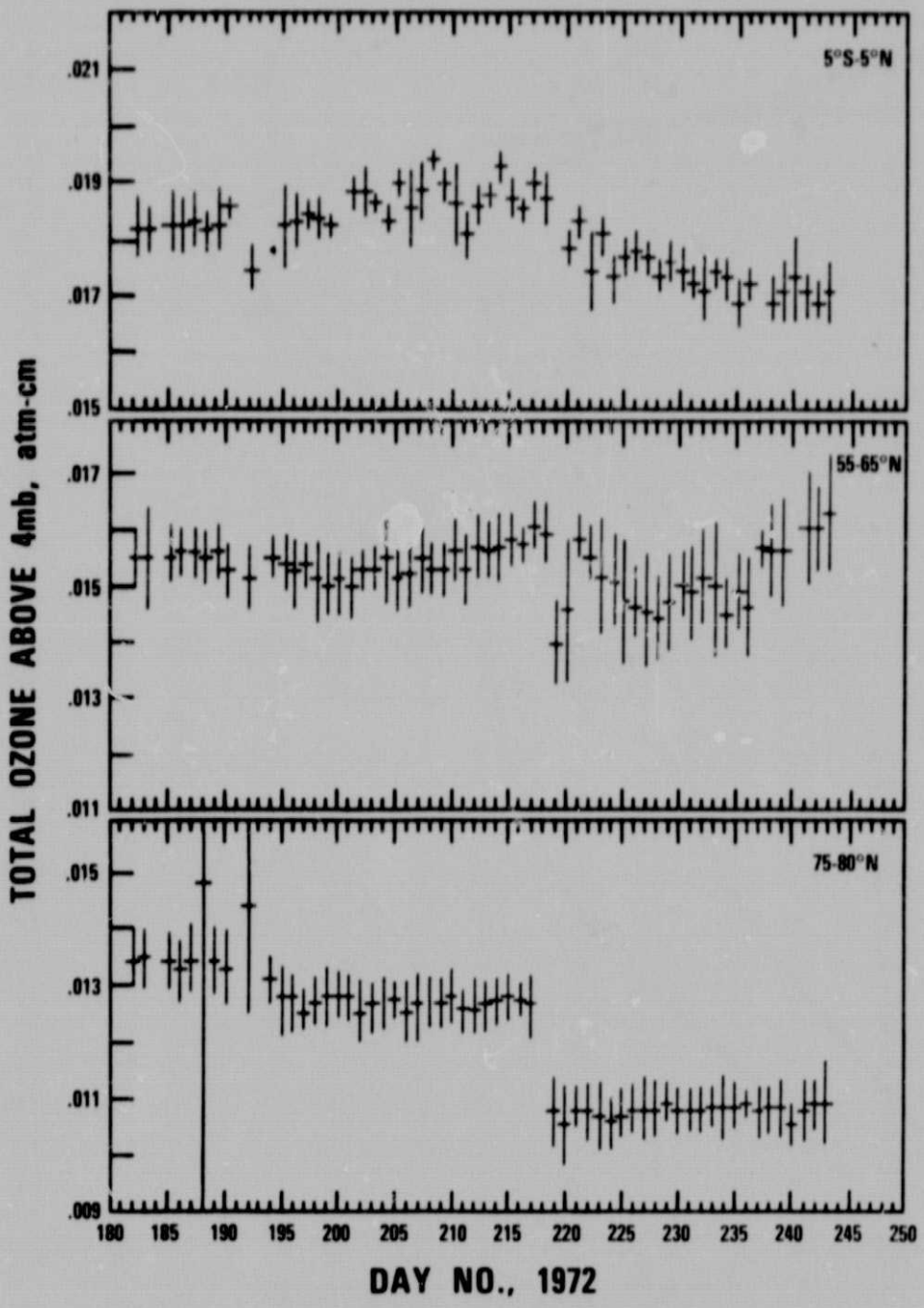
14. The observations may be an underestimate of the depletion. Artifacts in the data due to spacecraft pitch errors show positive biases increasing with time at the upper levels. As a result the ozone appears to increase above 1.1 mb at 8 days and above 1.3 mb at +19 days.

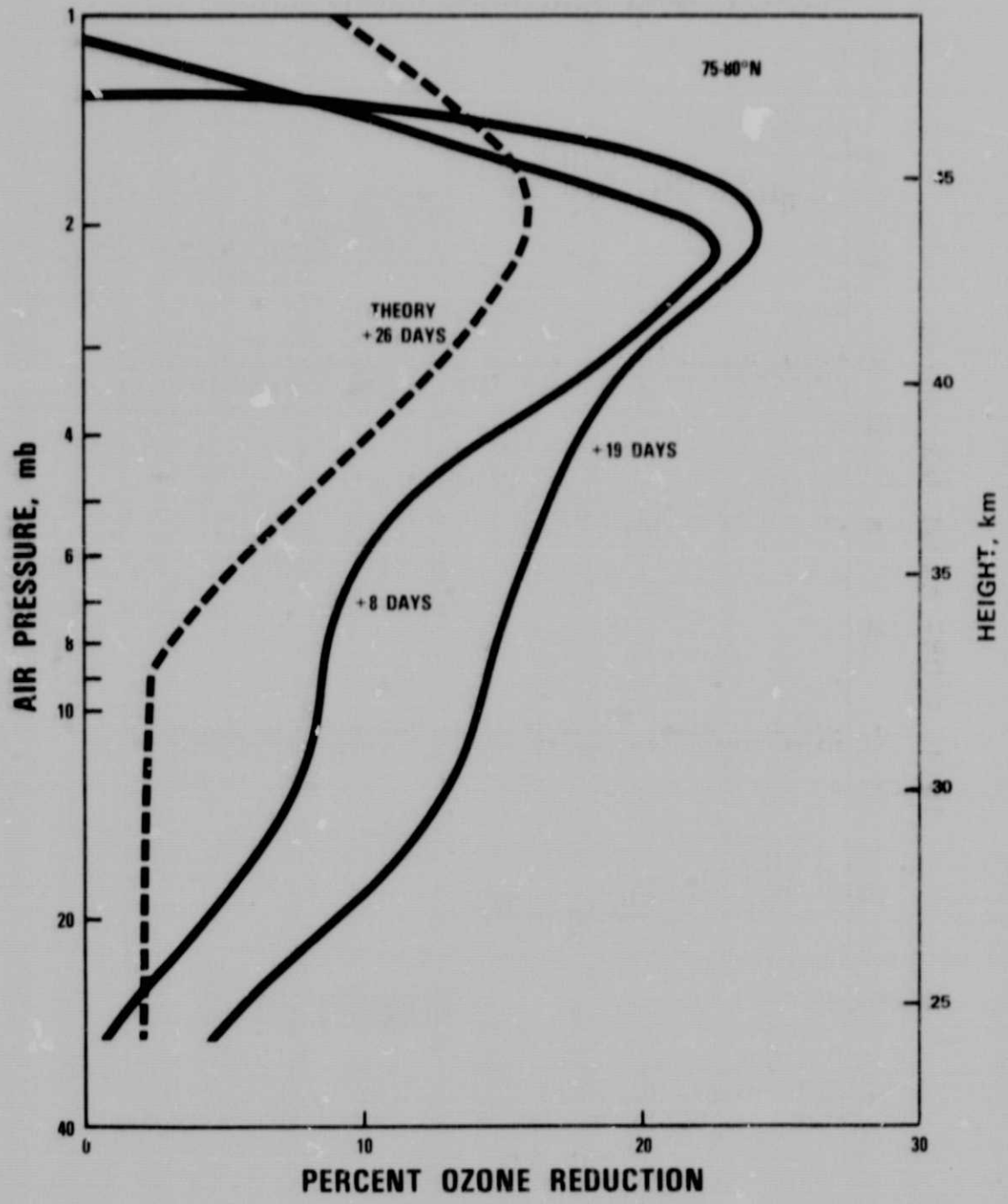
15. The ozone above 1 mb exhibited a perturbation which disappeared a few days after the event, in accordance with a short photochemical removal time for an HO_x deposition.

FIGURE CAPTIONS

Figure 1. Zonally-averaged total ozone above the 4 mb pressure surface for equatorial, middle and high latitudes during July and August 1972. The solar proton event occurred on August 4 (Day 217).

Figure 2. Percentage decrease of ozone partial pressure vs. air pressure derived from the average of the seven days before August 4, 1972, and seven day periods centered on 8 and 19 days after the solar proton event (solid lines). The dashed line is a calculation of the reduction for September 1, 1972, due to the catalytic effect of solar proton-produced nitric oxide.


5°S-5°N
55-65°N
75-80°N
.021
.019
.017
.015
.013
.011
.009
180
185
190
195
200
205
210
215
220
225
230
235
240
245
250
TOTAL OZONE ABOVE 4mb, atm-cm
DAY NO., 1972


75-80°N
THEORY
+26 DAYS
+19 DAYS
+8 DAYS
AIR PRESSURE, mb
HEIGHT, km
PERCENT OZONE REDUCTION
1
2
4
6
8
10
20
40
0
10
20
30
35
40
35
30
25