# Studies of the Observed and Theoretical Variations of Atmospheric Ozone

Final Report on NASA Grant NSG-5153

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## Final Report: NSG-5153

This is a final report of our research activities carried out under NASA Grant NSG-5153. During the thirteen years covered by the report (1 January 1977–30 June 1990), our grant research dealt with four main related topics:

1. distributions of total and upper atmospheric ozone and their time and space variations;
2. observed and theoretical models of the QBO ozone variation;
3. radiative processes in the upper atmosphere; and
4. relations between ozone and solar variations.

The results of these studies were published in twenty-three papers: twelve papers covered topic 1, four papers each covered topics 2 and 4, and three papers covered topic 3. (A complete reference list of publications describing the results of our research supported by grant funds is given below as Appendix A.) In addition, twenty-two papers discussing the results of grant research were presented at various symposia and workshops. Grant funds were used to support, partially or fully, six graduate-degree students and one postdoctoral research associate (a list of students supported by our NASA grant is given in Appendix B). The following information briefly summarizes the principal results of our research funded by NSG-5153.

## 1. Distribution and Variation of Total and Upper Atmospheric Ozone

Total-column ozone has been measured using Dobson-type instruments since 1925, but a quasi-global distribution of observing stations started only with the advent of the IGY in 1957. Currently, there are almost 100 stations taking routine total-ozone observations, principally using Dobson-type instruments, although some observations are based on optical filter measurements. The global-network observations are limited to land-based stations. Satellite measurements of total ozone started at the end of the 1960s and are derived from instruments on different satellites. They have the distinct and important advantage of providing almost continuous near-global distribution. Upper air ozone observations have been made from ground-based (Umkehr) observations principally since 1957, balloon-borne sounders and rocket measurements since the early 1960s, and satellite observations since early 1970. Details of the different ozone-measuring techniques were described in a number of published papers (2, 12, and 17).* The average observed global total-ozone distribution was also discussed in these papers, and in (6).

*Numbered papers are listed in Appendix A.

The long-term annual average distributions of total ozone as determined from ground-based and satellite-derived observations are in general mutual agreement despite the difference in time and space coverage of the observing systems. Analyses of the latitude, longitude, and seasonal distributions derived from the two systems of ozone measurements indicated significant similarity in the location and time variations of the north and south subpolar maxima and the equatorial ozone minimum. This is of particular interest since it permits some extrapolation back to the period when only ground-based data were available. As an example, we were able to show that even before the TOMS documentation, it was evident that an average total-ozone trough existed during September–October at latitudes south of about 65°S (17). However, a study of the geographic bias in determining average variations of total ozone from ground-based observations (10) indicated that although the absolute bias may be small (~1%), the rms difference could be as large as 2.5% where there are few ground-based stations (i.e., lat 40–50° S). Analysis of the trend of total ozone during the decade of the 1960s indicated a tendency for an increase of the order of 10% per decade in the Northern Hemisphere, but little significant change in the Southern Hemisphere with no obvious relation to the solar cycle, nuclear explosions, or volcanic
activity (1, 3). Indeed, an extension of this analysis covering 1958–1980 (17) showed that the strong total-ozone increase in the Northern Hemisphere detected earlier did not persist through the following decade.

Our analysis of the vertical ozone distribution was based on ozonesonde, Umkehr, and satellite observations. Ozonesonde data are most representative for ozone concentrations in the troposphere and lower stratosphere, Umkehr observations are particularly useful in the lower, middle, and upper stratosphere, and satellite data provide best results in the middle and upper stratosphere and lower mesosphere. The first two methods have long periods of records, but are severely limited by skeletal geographic distribution. Continuous satellite observations, on the other hand, are available for limited periods [i.e., slightly more than 1 year for OGO-4 (11) up to about 10 years for SBUV observations], but they do provide information that is generally quasi-global. We have shown that where the observations from these different techniques overlapped in time and space, the derived results tend to agree with each other quite well (17).

The global distribution of the ozone concentration in the troposphere, stratosphere, and lower mesosphere, including seasonal variations and hemispheric differences, was discussed in a number of our publications (2, 11, 12, 15, 17, 20, 21). Analyses of satellite-derived data were based largely on observations from the OGO-4, BUV, and SBUV systems. It was shown that in the upper troposphere and lower stratosphere, seasonal and longitudinal variations are quite pronounced at middle and high latitudes with ozone ridges during spring near the eastern continental coasts. Seasonal and longitudinal variations are relatively small in the subtropics. The long-term average pressure/latitude ozone mixing ratio for January and July indicated a maximum mixing ratio of about 16 ppmm near the equator at a level of 5–6 mb. The center of ozone maximum shifts from just south of the equator in January to just north in July. Seasonal variations of the equatorial maximum are in

response to seasonal variation of direct solar irradiance as it affects the maximum production of odd-oxygen species. The axis of ozone maximum tilts upward with increasing latitude during winter in each hemisphere.

Periodic (annual and semi-annual) variations were also calculated mainly from the BUV and SBUV observations (15, 21). The annual amplitude was computed to be largest in the subpolar upper stratosphere with a phase of maximum in winter. The maximum amplitudes at these heights and latitudes were larger by almost 50% in the Southern Hemisphere, a result of a combination of inverse temperature-dependent photochemistry and stronger slant-wise upward advection in the southern subpolar upper stratosphere. Other annual amplitude maxima are found in the subpolar regions near 10 and 40 mb. Dominant semi-annual variations were found at the equator at about 3–6 mb and slightly higher at subpolar latitudes. Hemispheric differences of time and longitude variations in the stratosphere and mesosphere were calculated from eight years of SBUV observations (20). These differences are evident mostly at the upper (1 mb) and lower (30 mb) levels where, as mentioned above, temperature-dependent photochemistry and quasi-horizontal advection (particularly in the lower stratosphere) are most effective. The longitude variations reflect, in large part, differences in the nature and form of the underlying boundary surface that, in turn, affect hemispheric differences in thermal structure and circulation characteristics of the stratosphere. Seasonal changes of the zonal variations (21) indicated that waves 1 and 3 are the most significant and are strongest during Northern Hemisphere winter at subpolar latitudes. A feature of the averaged zonal harmonic is the presence of two strong wave 1 oscillations at 10 and 40 mb over the tropics. These seem to persist during all seasons, but are strongest in July over the equator. There is a phase difference of about 180° between the lower- and upper-level wave 1. We had earlier pointed out (19) that at about 20 mb over the equator, the level of minimum amplitude of wave 1, the effects of vertical advection and temperature-dependent chemistry tend to cancel each other.

Some insight to the causes of the annual and semi-annual ozone variation can be provided by comparisons of the observed time, height, and latitude results with those derived from photochemical models. Where the observed and photochemical model distributions agree, analysis of the relative significance of transport and different photochemical terms in the model computation can indicate the basic mechanism responsible for the observed variations. The results of such an analysis (22) showed that in the low to middle stratosphere at subpolar latitudes, ozone abundances increase in winter because of transport from equatorial regions of production and decrease in summer because of chemical destruction. In the upper stratosphere, the annual ozone variation chiefly results from the annual temperature variation.

## 2. Observed and Theoretical Models of the QBO Ozone Variation

A quasi-biennial oscillation (QBO) of equatorial and subtropical total and lower and middle stratospheric ozone has been known since the early 1960s and 1970s. We used our previous analyses of monthly global distribution of total ozone and satellite-derived stratospheric ozone along with tropical stratospheric (50 mb) wind data to study the relation of the observed latitude-dependent total and stratospheric ozone QBO to the tropical stratospheric zonal wind oscillation (9, 14). Statistical spectra of the ozone variation showed maximum energy at about 28 months. A cross-spectrum analysis between monthly ozone and tropical stratospheric winds was performed at 10° latitude intervals to determine the coherence and phase lag between the ozone and wind data. The strongest coherence, at the 95% significance level, was found just north of the equator and at subpolar regions by using both ground-based and satellite-derived total-ozone values. The phase lag (west-wind maximum preceding ozone maximum) was found to be near zero at the equator and increased to about 60° at subpolar latitudes.

Theoretical studies using one- and two-dimensional models (19, 18) of the tropical and subtropical stratospheric ozone QBO were developed for a coupled radiative/photochemical/dynamic system. The results of these models showed quite good agreement in the height interval 20–35 km. The models were based on assumed zonal wind profiles to derive vertical wind, temperature, and ozone variations. In agreement with observations, the derived ozone QBO amplitude has a major maximum near 22 km and a secondary maximum at about 32 km. The phase of the 28-month ozone oscillation moves forward at a rate of slightly more than one month $km^{-1}$ between 35 and 29 km. It then experiences an abrupt shift of almost 12 months between 29 and 27 km and then advances again at an average rate of slightly more than one month $km^{-1}$.

The derived ozone stratospheric QBO amplitude decreases with latitude to a minimum at about 15° and then increases to a secondary maximum over the subtropics. This result was also in agreement with the satellite-derived values. The rapid phase increase occurs at a slightly higher level in the model than is observed. A diagnostic study in the model calculations showed clearly that the lower-layer major ozone QBO maximum was produced by oscillations of the vertical wind component. The upper-level maximum was the result of temperature-dependent photochemistry. The phase-lag dependence indicates the relative importance of the two different processes on the time-dependent ozone variation.

## 3. Radiative Processes in the Upper Atmosphere

The distribution of radiative energy sources and sinks helps to determine the thermal structure and modes of interaction involving photochemical and dynamic processes in the atmosphere. The important processes of concern in evaluating radiative heating and cooling in the Earth's atmosphere were briefly described (5). Calculations were made of the height/latitude/season distributions of the radiative

energy sources and sinks for the principal radiatively active gases in the stratosphere and mesosphere (7, 8). The results indicated that the composite, globally averaged net heating or cooling due to radiative processes was a slight warming ($< 1°$ day$^{-1}$) at about 20 km, radiative equilibrium at 25–45 km, and heating in the interval 45–80 km. The net radiative heating and cooling distribution has, of course, important latitude and season variations. The maximum heating occurs at high latitudes during summer at about 50 km. The maximum cooling was calculated to occur at high latitudes during winter at about 60 km. Details of the height/latitude distribution of the different computed heating and cooling rates due to the different atmospheric constituents are given in (7).

## 4. Relations Between Ozone and Solar Variations

From the onset of NASA grant NSG-5153 the subject of the response of atmospheric ozone to solar variability has always been a candidate problem for our investigation. Our main concern was to attempt to find a clear relationship between total or stratospheric ozone and solar UV variations on time scales longer than the solar rotation period (27 days). A study of the association between solar variability and total ozone (4) failed to show any significant statistical relationship between the two. However, in a later study (13) the results of an analysis of the association between satellite-derived stratospheric ozone and solar proxy data ($F_{10}$) suggested that there may very well be a positive response in the tropical stratosphere at a height of about 45–50 km. Adequate radiative/photochemical/dynamic models used to simulate the solar-ozone association discussed above require realistic input of the amplitude and phase of the principal solar UV forcing functions. We have made use of the Solar Mesosphere Explorer irradiance observations to determine the amplitudes of the UV irradiance variations for solar-rotation and solar-cycle periods (16). We also noted that there is an amplitude variation of the solar-rotation oscillation to produce an irradiance period of about 260–280 days. By comparing the SME UV irradiance values with the ACRIM/ERB solar constant observations during the decline phase of solar cycle 21, we were able to show that near-UV variability (200–300 nm) contributes about 30% -35% of the observed solar-constant decrease (23).

We are currently making use of the observed SME data as discussed above to model the ozone response in the stratosphere and mesosphere to solar variability at periods longer that 27 days. This ongoing study is being funded through the NASA/UARS program.

## APPENDIX A

### Publications of Research at Least Partially Supported Under Grant NSG 5153


1. London, J., 1978: Long period time variations of ozone in the lower stratosphere, Proceedings, 4th Joint Conference on Sensing of Environmental Pollutants, American Chemical Society, 677–680.

2. London, J., 1978: The distribution of atmospheric ozone and how it is measured, Air Quality Meteorology and Atmospheric Ozone, ASTM STP 653, A. L. Morris and R. C. Barras, eds., American Society for Testing and Materials, 339–364.

3. London, J. and S. J. Oltmans, 1978: The global distribution of long–term ozone variations during the period 1957-1975, *PAGEOPH,* **117,** (1978/79), Birkhauser Verlag, 345–354.

4. London, J. and C. A. Reber, 1979: Solar activity and total atmospheric ozone, *Geophys. Res. Lett.,* **6,** November 1979, 869–972.

5. London, J., 1980: Radiation problems in the earth's atmosphere, *Journal de Physique,* **41,** April 1980, 295–303.

6. London, J., 1980: The observed distribution and variations of total ozone, Proceedings of the NATO Advanced Study Institute on Atmospheric Ozone: Its Variation and Human Influences, Report No. FAA–EE–80–20, 31-44.

7. London, J., 1980: Radiative energy sources and sinks in the stratosphere, Proceedings of the NATO Advanced Study Institute on Atmospheric Ozone: Its Variation and Human Influences, Report No. FAA–EE–80–20, 703-721.

8. London, J. and G. G. Bjarnason, 1980: Zonally averaged heating rates in the stratosphere and mesosphere, Volume of extended abstracts, International Radiation Symposium, Fort Collins, CO., August 1980, 1–4.

9. Oltmans, S. J., and J. London, 1981: The quasi-biennial oscillation (QBO) in atmospheric ozone, Proceedings of the Quadrennial International Ozone Symposium, J. London, ed., Boulder, CO, August 1980, 397–405.

10. London, J. and X. Ling, 1981: The geographic bias in determining average variations of total ozone from ground-based observations, J. London, ed., Proceedings of the Quadrennial International Ozone Symposium, Boulder, CO, August 1980, 337–339.

11. Clayson, M. H., J. London and G. P. Anderson, 1981: The global distribution of stratospheric ozone from OGO–4 BUV observations, Proceedings of the Quadrennial International Ozone Symposium, J. London, ed., Boulder, CO, August 1980, 558–564.

12. London, J. and J. Angell, 1982: The observed distribution of ozone and its variations, *Man and Stratospheric Ozone,* **Vol. 1,** *Ozone and Perturbations,* F. A. Bower and R. B. Ward, eds., CRC Press, Inc., 7–42.

13. London, J., 1982: Satellite observed ozone variations and solar activity, Proceedings: International Conference on Environmental Pollution, September 1981, Thessaloniki, Greece, 799–810.

14. Oltmans, S. J., and J. London, 1982: The quasi-biennial oscillation in atmospheric ozone, *J. Geophys. Res.,* **87,** 8981–8989.

15. London, J., 1983: Periodic and aperiodic ozone variations in the middle and upper stratosphere, *Adv. Space Res.,* **2,** 201–204.

16. London, J., G. G. Bjarnason, and G. J. Rottman, 1984: 18 months of UV irradiance observations from the Solar Mesosphere Explorer, *Geophys. Res. Lett.*, **11**, 54–56.

17. London, J., 1985: The observed distribution of atmospheric ozone and its variations, in *Ozone in the Free Atmosphere,* R. C. Whitten and S. S. Prasad, eds., New York: Van Nostrand Reinhold Co., 11–80.

18. Ling, X., and J. London, 1985: A theoretical study of the quasi-biennial oscillation in the tropical stratosphere, in *Atmospheric Ozone,* Hampton: Deepak Publishing Co., 53–58.

19. Ling, X., and J. London, 1986: The quasi-biennial oscillation of ozone in the tropical middle stratosphere: a one-dimensional model, *J. Atmos. Sci.*, **43**, 3122-3137.

20. London, J., and L. Perliski, 1988: Hemispheric differences in observed stratospheric ozone, *Proceedings of the Quadrennial Ozone Symposium—88,* R. Bojkov and P. Fabian, eds., Hampton, VA: A. Deepak Publ. (in press).

21. Perliski, L. M., and J. London, 1989: Satellite observed long–term averaged seasonal and spatial ozone variations in the stratosphere, *Planet. and Space Science*, **37**, 1509–1526.

22. Perliski, L. M., S. Solomon, J. London, 1989: On the interpretation of seasonal variations of stratospheric ozone, *Planet. and Space Science*, **37**, 1527–1538.

23. London, J., J. Pap, and G. J. Rottman, 1989: Observed solar near UV variability: a contribution to variations of the solar constant, *MAP Handbook*, **29**, *Proceedings of the IAGA Symposium on Solar Activity Forcing of the Middle Atmosphere,* J. Lastovicka (ed.), available from SCOSTEP Secretariat, U. of Illinois, Urbana, IL 61801, 9–12.

## APPENDIX B

### Students At Least Partially Supported By Grant Funds

Ming-san Zhang M.S. 1983

Joseph Coughlin M.S. 1984

Mark Clayson M.S. 1984

Xiu-de Ling Ph.D. 1984

Gudmunder Bjornason Ph.D. 1987

Lori M. Perliski M.S. 1989 (Ph.D. in progress)

### Postdoctoral Research Associate

Dr. Judit Pap 1989