THE GLOBAL CONSEQUENCES OF INCREASING TROPOSPHERIC OZONE CONCENTRATIONS

Jack Fishman
Atmospheric Sciences Division (Mail Stop 401A)
NASA Langley Research Center
Hampton, Virginia 23665-5225

Recent analyses of long-term records of tropspheric ozone measurements in the Northern Hemisphere suggest that it is increasing at a rate of 1 to 2 percent per year. Because of this, it is argued that the amount of atmsopheric warming due to increasing troposphreic ozone is comparable to, or possibly even greater than, the amount of warming due to the increase of carbon dioxide. Unlike all other climatically important trace gases, ozone is toxic, and increases in its concentration will result in serious environmental damage, as well as impairment of human health.

February 1989
Submitted to Nature

Introduction.

The recent study by Hansen et al. (ref. 1) presents a detailed set of calculations illustrating the global effects of increasing concentrations of trace gases in the atmosphere that are primarily the result of anthropogenic activity. In testimony before Congress, Hansen presented a scientific scenario in which he proposed that signs of the Greenhouse Effect are already present, in that four of the warmest years on record have occurred in the 1980's (ref. 2). Much of the basis for this scenario was concluded from the Hansen et al. (ref. 1) general circulation model (GCM) study. In that model study, however, very little attention was paid to the reported increases in tropospheric ozone that have been observed over the past two decades[3-8]. When these increases are considered, it can be shown that the contribution to global warming due to the increase of tropospheric ozone is at least comparable to, and possibly even greater than, the increase in temperature that has resulted from the increase in carbon dioxide over the same period of time.

Although the problem of ozone pollution has traditionally been the concern of urban scale air-quality research where very high concentrations have been known to cause plant and material damage, as well as being an eye and respiratory irritant for humans, more recent studies now find that ozone concentrations that are often found in nonurban areas are causing environmental and health damage. Thus, the impact of ozone formation that was once

the subject of only urban or regional scale photochemical studies now appears to have important repercussions on scales that impact the entire Northern Hemisphere.

Tropospheric Ozone as a Greenhouse Gas.

The importance of tropospheric ozone as a significant climatic trace gas was first recognized by Ramanathan and Dickinson[9]. Subsequently, Fishman et al.[10] presented a set of calculations using a radiative transfer model[9] which calculated the following results:

1. Because there is more ozone in the Northern Hemisphere than in the Southern Hemisphere, the difference in the temperatures of the two hemispheres due to this effect should be ~0.2° K;

2. If tropospheric ozone has already doubled since the onset of the Industrial Revolution (circa late 1800's), the temperatures have already increased by ~0.5° K; and

3. If the amount of tropospheric ozone increases from the present concentrations to twice as much in the future, then the surface temperature will increase by ~0.9° K. For comparison, a doubling of CO_2 by this model would result in an increase of 2-3° K.

Another way of looking at the last result would be that a doubling of tropospheric ozone is approximately 30-40 percent as effective in warming the atmosphere as would be a doubling of atmospheric CO_2.

Using a similar type of model, Hansen et al.[1] arrive at a conclusion similar to the results summarized in Fishman et al.[10] and other studies that used radiative-convective models to examine the comparative aspects due to increases of carbon dioxide, tropospheric ozone, and other radiatively important trace gases[11]. For example, Ramanathan et al.[11] calculate a

temperature increase of 0.71° K for a 32 percent rise in CO_2 by the year 2030; for an increase in tropospheric O_3 of 15 to 50 percent over the same period of time, they calculate a temperature rise of 0.1 to 0.4° K. Extrapolation of both scenarios to a doubling of each trace gas shows that a doubling of tropospheric ozone increases temperatures ~35 percent as much as a doubling of CO_2. A slightly different scenario for the year 2050 in Dickinson and Cicerone[12] yields a ratio of ~30 percent.

In the Hansen et al.[1] study, however, they assume that tropospheric ozone will increase by 25 percent in the same amount of time that is required to double CO_2 from 315 ppm (parts per million) to 630 ppm. They state that such a doubling of CO_2 would warm the surface by ~1.2° K, whereas a 25 percent increase in tropospheric ozone would result in a warming of ~0.1° K. To a first approximation, they would calculate that a doubling of tropospheric ozone would have an impact of ~0.4° K, or about one-third the impact of doubling CO_2, a result in good agreement with earlier independent calculations. Thus, to understand how the amount of warming due to the increase of tropospheric ozone compares with the amount of warming due to the increase of CO_2, we must compare the relative rates of increase of these two trace gases over the same period of time.

Recent Trends in Tropospheric Ozone.

One of the difficulties in determining the trend of tropospheric ozone is that its atmospheric lifetime is considerably shorter (on the order of a few days to several months) than the lifetime of CO_2 and all other greenhouse trace gases which have an atmospheric residence time of at least

several years. Therefore, considerably more analysis must go into the determination of such a trend since the inherent variability of tropospheric ozone measurements makes such data considerably more difficult to interpret. Ozone measurements are also subject to interference from the presence of other gases that are often concurrently present in polluted areas.

Determination of trends using long-term measurements has been a problem for several reasons. Most air-quality measurements are located near urban centers. In many cases, stations that were once considered "rural" when they were first established in the 1950's and 1960's have become engulfed in a metropolitan complex by the 1980's. Measurements above the boundary layer using ozonesondes should, in principle, measure concentrations that are fairly representative of the concentrations of the background troposphere. However, because of the pump-efficiency problems when ozonesondes reached stratospheric altitudes, many of the soundings were corrected by normalizing them with ground based spectroscopic measurements of total ozone. Such a procedure often introduced artificial corrections to the concentrations measured in the troposphere. In addition, many of the long-term ozonesonde measurements (which were started primarily in the early 1960's to assist in the monitoring of radioactive fallout from the stratosphere) switched measurement techniques in the mid-1960's. This switch of instrumentation introduced different responses to contamination of the sensors by local pollution. For further details, refer to Logan[7].

Because of the relatively short atmospheric residence time, another difficulty in the analysis of tropospheric ozone arises when observations from a few selected stations are extrapolated to the globe. For the present

study, the analysis is confined to the Northern Hemisphere, and it should be noted that no increase in the concentration of tropospheric ozone has been observed for the very limited set of observations in the Southern Hemisphere[3]. These hemispheric trends are consistent with the original hypothesis of Fishman and Crutzen[13], which stated that tropospheric ozone concentrations should have increased considerably in the Northern Hemisphere since the onset of the Industrial Revolution.

Table 1 summarizes a series of independent studies that have been conducted recently to calculate the rate of increase of tropospheric ozone in the Northern Hemisphere over the past 15-30 years. All of these analyses suggest that ozone has increased throughout the troposphere during this time. Many of the data sets are totally independent from each other and they include observations from locations that may have been impacted by regional-scale increases (such as the stations in East Germany and Switzerland) as well as stations that were carefully chosen to be representative of the background atmosphere (e.g., Mauna Loa and Barrow). They include both surface measurements and free tropospheric observations obtained from ozonesonde data. In summary, they conclude that tropospheric ozone has increased at an annual rate of between 0.8 and 2.3 percent per year over the past 20 years or so. For the purposes of the discussion presented in this study, it will be assumed that tropospheric ozone has increased between 1 and 2 percent per year for the period between 1965 and 1985. Such an increase translates to an increase of between 22 and 48 percent between 1965 and 1985.

Between 1965 and 1985, Hansen et al.[1] utilize an increase of ~6 percent in the concentration of CO_2 for the basis of their calculations. Incorporating the range of uncertainty into the relative trends of tropospheric O_3 and CO_2, as well as the uncertainty of their relative contributions to the greenhouse effect, this simple calculation shows that tropospheric ozone has an impact that is at least comparable to the impact of the temperature increase due to increasing CO_2 concentrations over the same period of time. At the other extreme, it can be argued that the increase in tropospheric ozone has warmed the atmosphere 1.5 times as much as the corresponding increase of CO_2 between 1965 and 1985. In either case, it is clear that the contribution of the increase of tropospheric ozone to the climate question cannot be ignored.

Figure 1 summarizes the relative impact of the increases of several trace gases in the troposphere over the 20 year[1]. The number over each of the bars indicates the percentage increase of the trace gas between 1965 and 1985. The difference between this plot and the one that appeared in Hansen et al.[1] is that we have assumed that tropospheric ozone has increased by between 22 and 48 percent between 1965 and 1985; the hatched area represents the calculated range of the warming depending on whether a 1 percent per year increase, or a 2 percent per year is assumed.

An important difference between the impact of increased concentrations of tropospheric ozone on climate and the impact of increased CO_2 concentrations on climate is the fact that increases in tropospheric ozone in the upper troposphere have more of an effect than increases on tropospheric

ozone at the surface. The preceding discussion has been based on calculations that assume that ozone concentrations throughout the entire troposphere are increasing uniformly at the same rate. From analyses of ozonesonde data, both Logan[7] and Bojkov[6] conclude that the increases in tropospheric ozone are taking place throughout the troposphere although these increases are somewhat less in the upper troposphere than at the surface. For Hohenpeissenberg, which is probably the best data set for northern mid-latitudes because of the large number of measurements that went into the analysis, Logan reports a trend of 2.6 $\pm$ 0.7 percent per year near the surface, 1.9 $\pm$ 0.3 percent per year for the layer between 700 mb and 500 mb, and 0.8 $\pm$ 0.8 percent per year at 300 mb. With the exception of Goose Bay, which is the only station analyzed by Logan[7] that shows a negative trend for tropospheric ozone, all eight other stations in the Northern Hemisphere show increases of 0.7 $\pm$ 0.5 percent to 1.7 $\pm$ 0.7 percent per year in the free troposphere. Similarly, Oltmans and Komhyr[3] conclude from a comparison of ozonesonde data taken from nearby Hilo, that their analysis of the Mauna Loa data at the surface is representative of the ozone behavior in the lower free troposphere.

Synergistic Effects.

Increases in tropospheric ozone have important implications for the interpretation of the trend of stratospheric ozone depletion since it is clear that any decrease in the Northern Hemisphere stratosphere will be underestimated from an analysis of total ozone measurements from both ground-based instrumentation and from satellites[14]. Since tropospheric

ozone comprises 10-15 percent of the total ozone amount in the atmosphere,[7,15,16] an increase of 1-2 percent per year in the troposphere would cause a downward trend in the stratosphere to be underestimated by 0.1-0.3 percent per year. This factor may account for some, or much of the finding that total ozone amounts have decreased by 2.9 percent between 0^{o} and 53^{o} S, but by only 1.8^{o} percent between 0^{o} and 53^{o} N for the period 1978-1987.

Furthermore, if emissions of the precursors to ozone formation continue to rise, the concurrent decrease in stratospheric ozone has a positive feedback on the rate of photochemical ozone production in the troposphere by allowing more ultraviolet radiation to reach the lower troposphere. Increased ultraviolet radiation between 300 and 320 nm will directly enhance the rate of formation of the OH radical, which is the means by which most oxidation sequences in the troposphere are initiated[17].

Unpleasant Consequences.

Unlike all other greenhouse gases (primarily carbon dioxide, methane, chlorofluorocarbons, and nitrous oxide) that may contribute to global warming, ozone has one unique characteristic that sets it apart: ozone is a poisonous gas. Because of its toxicity, it is already estimated that ozone causes more than 1 billion dollars each year in crop damage[18] in the United States. Recent studies have also shown that much of the "Forest Dieback" common to the Schwarzwalde in Germany can be linked to the recent increases in tropospheric ozone[19]; similar symptoms are currently being found in the

eastern United States, and are likewise being attributed to long-term increases in tropospheric ozone concentrations[19]. Damage to forests would be even more acute when associated with the likely possibilty that the frequency of drought conditions will also be more common in many forested regions as the climate warms.

Another synergistic effect may also result from the fact that elevated ozone concentrations directly affect the growth rate of trees[20]. For example, sweetgum (Liquidambar styraciflua L.) seedlings exposed to ambient concentrations of 0.10 ppm for 6 hours a day over a 28-day period exhibited a 29 percent reduction in growth when compared with a control group for which ozone was removed from the air. Numerous studies have shown that the rate of photosynthesis of many common trees likewise is slowed when ambient ozone concentrations are higher[21]. With less foliar biomass, the rate of uptake of CO_2 by forest ecosystems diminishes, resulting in a further build-up in carbon dioxide.

Perhaps most ominous is the fact that scientists have recently found that human health is adversely impacted at the ozone concentrations commonly measured throughout much of the United States during the summer[22,23]. For example, studies have shown that pulmonary function is significantly reduced in healthy children and adults in rural areas, even at ozone concentrations that are below the standards established by the United States Environmenal Protection Agency. The detrimental aspects of increased tropospheric ozone on human health becomes even more acute when such increases in the amount of ozone in the air we breathe are concurrent with warmer temperatures.

Long-term Trends Since the 1800's.

Ozone was discovered in 1839 by C.F. Schônbein. Soon after its discovery, it was measured at many locations around the world. A review of many of these early measurements during the latter half of the nineteenth century is given by Bojkov[24], where he summarizes that the concentrations ~100 years ago were considerably lower than what they are today. Absolute quantitative estimates of the concentrations obtained by the instruments used to measure ozone at that time are, however, complicated by possible flaws in the calibration of these instruments and by potential interferences from trace gases other than ozone (primarily SO_2).

Because of its reactivity, ozone cannot be stored for long periods of time, nor can information about its concentration be obtained from ice core samples. Examination of historic spectra, such as was done for atmospheric methane[25], likewise cannot be used to obtain information about tropospheric ozone since most ozone is in the stratosphere. Thus, at the present time, it appears that the best way to determine the absolute concentration of ozone before the "modern era" is to carefully interpret the older records that utilized the Schônbein technique.

Probably the most comprehensive set of early ozone data was obtained at the Meteorological Observatory of Montsouris at the south perimeter of Paris where continuous measurements were obtained for 34 years beginning in 1876. The Montsouris apparatus was reconstructed[26] and its performance was compared with modern instrumentation. The records from the Meteorological Observatory were then analyzed to screen out the times when pollution from the city likely contaminated the measurements. Volz and Kley[26] conclude

that air that was representative of background (rural) conditions around Paris contained ~0.010 ppm of ozone, and that this concentration is accurate to within 20 percent. They furthermore concluded that there was no significant trend in that 34-year data set.

Comparing those measurements with present-day surface measurements in rural Europe, we conclude that today's concentrations of 0.020-0.030 ppm under comparable conditions represents an increase in tropospheric ozone of between 100 and 200 percent. During the same time period, atmospheric CO_2 concentrations have increased by ~25 percent. Recalling that Fishman et al.[10] calculated a temperature increase of ~0.5° K using the model of Ramanathan and Dickinson[9] if present day levels of tropospheric ozone have already reached concentrations that are twice as large as those that were present before the onset of the Industrial Revolution, the same model calculates a warming of ~0.6° K for a 25 percent increase of CO_2. If tropospheric ozone concentrations have tripled over that time period, then the atmosphere would have warmed by ~0.7° K. Thus, the increase in temperature since the turn of the century due to higher concentrations of tropospheric ozone is comparable to the warming due to carbon dioxide.

Extrapolation to a Global Increase

Even if the Montsouris data accurately represent the surface concentration of ozone over Europe ~100 years ago, the question remains as to the representativeness of these measurements to other parts of the Northern Hemisphere. At the present time, there is no way to resolve this question.

On the other hand, some recent field experiments[27-30] and analyses of satellite data sets to quantify the amount of ozone in the troposphere[31-33] have shown that significant quantities of ozone are also produced from biomass burning in the Tropics. For many of the trace gases that are the precursors for photochemical generation of tropospheric ozone, the magnitude of this tropical source is estimated to be comparable to their source strengths at northern mid-latitudes[34]. It is likely that this low-latitude source of tropospheric ozone has likewise increased considerably over the past few decades as slash-and-burn agricultural practices and deforestation have expanded to support the increasing population of these tropical nations (most of which are in the northern Tropics). The rate of population growth in these tropical countries is more than three times faster than the rate of growth of mid-latitude countries[35].

To determine the potential impact of tropospheric ozone on global climate with greater accuracy, it first becomes necessary to incorporate an accurate depiction of its spatial and seasonal behavior into the framework of a GCM. Such a task is considerably more difficult than the inclusion of all other climatically important trace gases that do not exhibit significant spatial gradients. Until such a model is available, it will be difficult to assess the climatic impact of increasing trospospheric ozone more quantitatively.

Irrespective of the climatic impact of increasing tropospheric ozone concentrations, the more significant aspect of increasing amounts of this gas is that it causes damage to living things on this planet. The ecological and potential human health aspect of this problem are just beginning to

be assessed. Tropospheric ozone may be the nonlinear component of the Global Change scenario that might be considerably more damaging to the environment and the well-being of humankind than either climatic warming or stratospheric ozone depletion.

The author thanks D. McKee of the U.S. EPA for providing many of the references used in this study. This paper has benefited from comments on earlier versions of this manuscript by P. Minnis, L. Callis and C. Rinsland of NASA Langley. I thank D. McKee, V. Ramanathan of the University of Chicago, J. Logan of Harvard University, S. Oltmans of NOAA, and R. Kalish of New Hampshire College for their stimulating discussions. This research has been supported in part by NASA's Global Trophspheric Chemistry Program.

TABLE 1.

Trends in Ozone in Long-Term Records

Altitude of Measurement	Location	Number of Stations	Period of Observation	Annual Rate of Increase	Reference
Surface (3400 m)	Mauna Loa (20° N)	1	1974-1984	1.4%	(3)
Surface	East Germany (50° - 55°N)	5	1972-1983	2.3%	(4)
Surface	Barrow (71° N)	1	1973-1983	0.8%	(3)
850 mb (sonde)	Switzerland (48° N)	2	1967-1986	2.0%	(5)
850 mb (sonde)	Barrow (71° N)	1	1973-1986	1.6%	(5)
1-12 km	Switzerland (48° N)	2	1967-1982	2.0%	(6)
700 mb-500 mb	32° N - 75° N	8	1968-1982 (9-13 years)	0.9%	(7)
800 mb-500 mb	32° N - 75° N	12	1966-1982 (10-16 years)	0.8%	(8)

Figure Caption

Figure 1. The increase in surface temperature is computed using observed increases of various trace gases between 1965 and 1985. The number above each of the bars represents the observed percentage increase of each trace gas over that 20-year period. For tropospheric ozone, the effects of a 1 percent per year increase (22 percent over 20 years) and a 2 percent per year increase (48 percent over 20 years) are shown by the solid bar, and the hatched area above it, respectively.

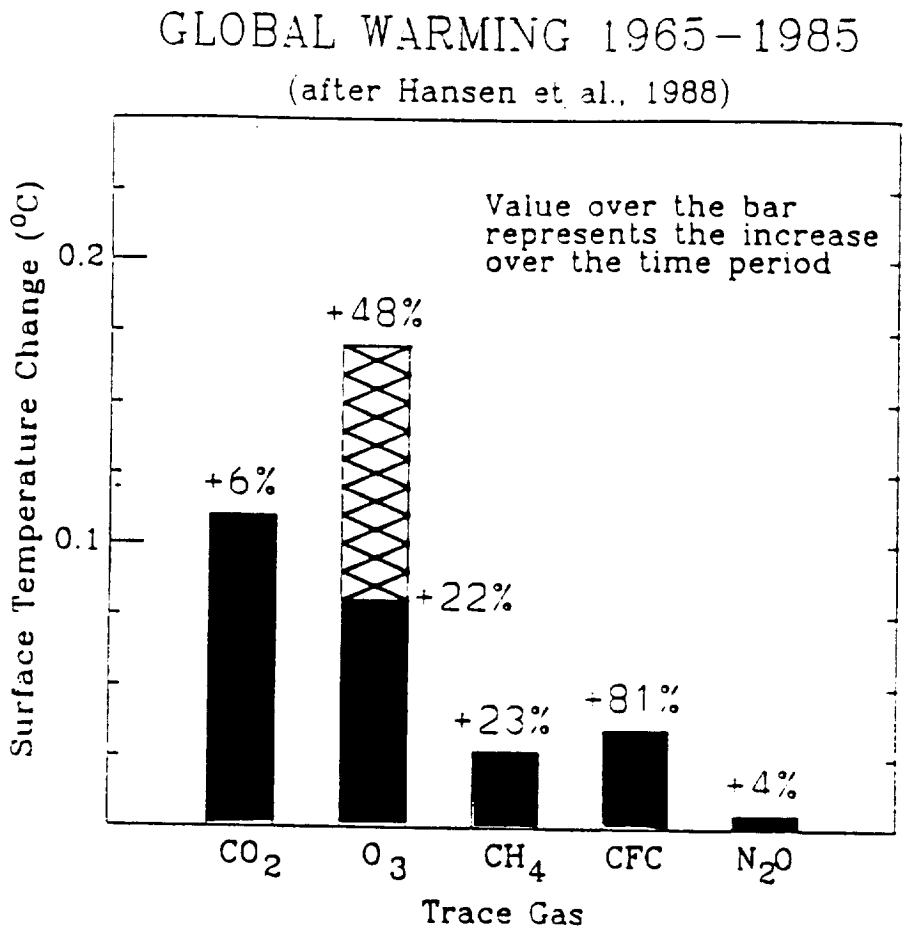

Figure 1. The increase in computed surface temperature is computed using observed increases of various trace gases between 1965 and 1985. The number above each of the bars represents the observed percentage increase of each trace gas over that 20-year period. For tropospheric ozone, the effects of a 1 percent per year increase (22 percent over 20 years) and a 2 percent per year increase (48 percent over 20 years) are shown by the solid bar, and the hatched area above it, respectively.

References

1. Hansen, J., Fung, I., Lacis, A., Rind, D., Lebedeff, R., Ruedy, R., & Stone, P., J. Geophys. Res., 93, 9341-9364 (1988).
2. Discover, 50-61 (October 1988).
3. Oltmans, S.J. & Komhyr W.D., J. Geophys. Res., 91, 5229-5236 (1986).
4. Feister, U. & Warmbt W., J. Atmos. Chem., 5, 1-22 (1987).
5. Bojkov R.D., in Tropospheric Ozone, Reqional and Global Scale Interactions, (ed. Isaksen, I.S.A) 83-96 (Reidel, Dordrecht, Netherlands, 1988).
6. Bojkov R.D., in Proc. WMO Conf. Observat. Atmos. Contamin. Vienna, 94-127 (WMO Environ. Rep. No. 16, Geneva, 1983).
7. Logan, J.A., J. Geophys. Res., 90, 10463-10482 (1985).
8. Tiao G.C., Reinsel, G.C., Pedrick, J.H., Allenby, G.M., Mateer, C.L., Miller, A.J., & DeLuisi, J.J., J. Geophys. Res., 91, 13121-13136 (1986).
9. Ramanathan, V. & Dickinson, R.E., J. Atmos. Sci., 36, 1084-1104 (1979).
10. Fishman J., Ramanathan, V., Crutzen, P.J. & Liu, S.C., Nature, 282, 818-820 (1979).
11. Ramanathan V., Cicerone, R.J., Singh, H.B., & Kiehl, J.T., J. Geophys. Res., 90, 5547-5566 (1985).
12. Dickinson, R.E. & Cicerone, R.J., Nature, 319, 109-112 (1986).
13. Fishman, J., & Crutzen, P.J., Nature, 274, 855-858 (1978)

14. Watson, R.T. et al., Present State of Knowledge of the Upper Atmosphere 1988: An Assessment Report, 201 pp. (NASA Headquarters, Washington, 1988).

15. London, J., in Ozone in the Free Atmosphere, (eds. Whitten R.C. & Prasad, S.S.) 11-80 (Van Nostrand Reinhold, New York, 1985).

16. Chatfield, R.B. & Harrison, H., J. Geophys. Res., 82, 5965-5976 (1977).

17. Crutzen, P.J., in Tropospheric Ozone: Regional and Global Scale Interactions (ed. Isaksen, I.S.A.) 3-32 (D. Reidel, Dordrecht, 1988).

18. Heck, W.W., Taylor, O.C., Adams, R., Bingham, G., Miller, J., Preston, E., & Weinstein,L., J. Air Poll. Contr. Assoc., 32, 353-361 (1982).

19. Prinz, B., Environment, 29, 11-37 (1987).

20. Pye, J.M., J. Environ. Qual., 17, 347-360 (1988).

21. Air Quality Criteria for Ozone and Other Photochemical Oxidants, EPA/600/8-84/020cF (Vol. 3),(U.S. EPA Environmental Criteria and Assessment Office, Research Triangle Park, NC, 1986).

22. Folinsbee L.J., McDonnell, W.F.,& Horstman, D.H., J. Air Poll. Contr. Assoc., 38, 28- 35 (1988).

23. D.M. Spektor et al., Am. Rev. Respir. Dis., 137 313-320 (1988).

24. Bojkov, R.D., J. Clim. Appl. Meteor., 25, 333-352 (1986).

25. Rinsland, C.P., Levine J.S. & Miles, T., Nature, 318, 245-248 (1985).

26. Volz, A. & Kley, D., Nature, 332, 240-242 (1988).

27. Delany, A.C., Haagenson, P., Walters, S., Wartburg, A., & Crutzen, P.J., J. Geophys. Res., 90, 2425-2429 (1985).

28. Crutzen, P.J., Delany, A.C., Greenberg, J., Haagenson, P., Heidt, L., Lueb, R., Pollock, W., Wartburg, A. & Zimmerman, P., J. Atmos. Chem., 2, 233-256 (1985).

29. Andreae M.O., et al., J. Geophys. Res., 93, 1509-1527 (1988).

30. Fishman, J. & Browell, E.V., Tellus, 38B, 393-407 (1988).

31. Fishman J., Minnis, P., & Reichle, Jr., H.G., J. Geophys. Res., 92, 14451-14465 (1986).

32. Fishman, J. & Larsen, J.C., J. Geophys. Res., 92, 6627-6634 (1987).

33. Fishman, J., in Tropospheric Ozone: Regional and Global Scale Interactions, (ed. Isaksen I.S.A.) 111-123 (D. Reidel, Dordrecht, 1988).

34. Logan, J.A., Prather, J.J., Wofsy, S.C. & McElroy, M.B., J. Geophys. Res., 86, 7210-7254 (1981).

35. Brown, L., State of the World, (Worldwide Institute, Washington, 1986).