

PROGRESS IN ENVIRONMENTAL SCIENCE



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Observing tropospheric ozone from space

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Abstract: The importance of tropospheric ozone embraces a spectrum of relevant scientific issues ranging from local environmental concerns, such as damage to the biosphere and human health, to those that impact global change questions, such as climate warming. From an observational perspective, the challenge is to determine the tropospheric ozone global distribution. Because its lifetime is short compared with other important greenhouse gases that have been monitored over the past several decades, the distribution of tropospheric ozone cannot be inferred from a relatively small set of monitoring stations. Therefore, the best way to obtain a true global picture is from the use of space-based instrumentation where important spatial gradients over vast ocean expanses and other uninhabited areas can be properly characterized. In this paper, the development of the capability to measure tropospheric ozone from space over the past 15 years is summarized. Research in the late 1980s successfully led to the determination of the climatology of tropospheric ozone as a function of season; more recently, the methodology has improved to the extent where regional air pollution episodes can be characterized. The most recent modifications now provide quasi-global (50°N to 50°S) maps on a daily basis. Such a data set would allow for the study of long-range (intercontinental) transport of air pollution and the quantification of how regional emissions feed into the global tropospheric ozone budget. Future measurement capabilities within this decade promise to offer the ability to provide concurrent maps of the precursors to the *in situ* formation of tropospheric ozone, from which the scientific community will gain unprecedented insight into the processes that control global tropospheric chemistry.

Key words: global pollution; satellite measurements; tropospheric ozone.

1 Introduction

The classic set of measurements of carbon dioxide from the Mauna Loa Observatory, initiated during the International Geophysical Year in 1957 and then meticulously carried out over the next four decades has provided the world with the most convincing evidence that human activity is affecting our planet in global proportions.

Over the last four decades, the Mauna Loa record has shown that CO₂ concentrations have increased from 315 ppmv (parts per million, by volume: mixing ratios of 10⁻⁶) to more than 375 ppmv (current data can be found at the web site: <http://www.mloserv.mlo.hawaii.gov/ccgml0.htm>). As scientific measurement capabilities became more sophisticated during the 1970s and 1980s, we now have the ability to make accurate measurements down to less than one part per trillion (mixing ratios of 10⁻¹²). As a result, we now realize that in addition to CO₂, concentrations of a number of other trace gases have increased considerably. It is now well documented that methane (CH₄) and nitrous oxide (N₂O), both of which, like CO₂, occur naturally in the atmosphere, as well as all man-made chlorofluorocarbon (CFC) compounds have increased considerably over the past 40 years. Examination of air bubbles trapped in ice cores generally show that the increase in the natural trace gases started last century with the onset of the Industrial Revolution. All of the above gases absorb radiation in the infrared portion of the electromagnetic spectrum, and increases in their concentrations will contribute to the Earth's natural greenhouse effect and thus to the possibility of global warming. All of the above greenhouse gases also have residence times in the atmosphere on the order of a decade or more, which means that measurements at one location on the planet will be similar to measurements anywhere else in the world since their long residence times allow global mixing to occur.

Another gas that increased considerably over the past century is tropospheric ozone (Volz and Kley, 1988; Marenco *et al.*, 1994). One of the difficulties in determining the trend of tropospheric ozone is that its atmospheric residence time is considerably shorter (on the order of a few days in the planetary boundary layer to several months in the upper troposphere) than the lifetimes of the trace gases mentioned above. A number of reliable monitoring sites at northern temperate latitudes indicate that tropospheric ozone concentrations are increasing at a rate of 1–3% per year. The increase, however, is quite variable, and there are some stations (e.g., American Samoa in the tropical South Pacific and Antarctica) where a significant increase over the past 20 years or so cannot be detected (Oltmans and Levy, 1994). Some climate-chemistry models predict that increases in tropospheric ozone over the next two decades will mean that ozone will become the second most important contributor to the enhanced greenhouse effect, after CO₂ (Pszenny and Brasseur, 1997).

Unlike all other greenhouse gases 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 US\$1 billion each year in crop damage in the USA (Tingey *et al.*, 1994). From a global perspective, Chameides *et al.* (1994) have hypothesized that decreased crop yields resulting from rising tropospheric ozone concentrations may contribute to global food shortages by 2025. 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 (Prinz, 1987); similar symptoms are currently occurring in the eastern USA and are likewise being attributed to long-term increases in tropospheric ozone concentrations. Observed decreased growth rate of forests may also be attributable to increasing tropospheric ozone concentrations (Pye, 1988).

Perhaps most ominous is the fact that a number of scientific studies have found that human health is adversely impacted at the ozone concentrations commonly measured throughout much of the USA during the summer (Horvath and McKee, 1994). In a

number of laboratory studies in the 1980s, it was shown that ozone concentrations representative of those found in many areas of the USA are sufficiently high to elicit measurable transient changes in lung function, respiratory symptoms and airway inflammation in healthy people engaged in normal outdoor exercise and recreational activities (Lippmann, 1989). Subsequent studies now suggest that chronic exposure to ozone concentrations commonly found in the USA (Galizia and Kinney, 1999; Kinney and Lippmann, 2000) and Australia (Jalaludin *et al.*, 2000) have an adverse effect on lung function. One of the US reports concludes that persons living for four or more years in regions with high levels of ozone and related copollutants are associated with both diminished lung function and more frequent reports of respiratory symptoms (Galizia and Kinney, 1999).

Elevated ozone concentrations are also associated with widespread burning practices in subtropical and tropical regions (Crutzen and Andreae, 1990), such as when residents in southeast Asia were encouraged to stay indoors in 1997 during the height of the burning. In addition to southeast Asia (Fujiwara *et al.*, 1999), high ozone concentrations associated with extensive biomass burning also plague South America and Africa (Kirchhoff and Marinho, 1994; Cros *et al.*, 1992). Thus, the importance of tropospheric ozone embraces many facets of relevant scientific issues, from local environmental problems to global change implications. The challenge from an observational perspective is to obtain the proper characterization of the global distribution of tropospheric ozone and then to determine how this distribution is being perturbed by anthropogenic activity.

One particularly critical aspect regarding ozone centers on acquiring a good knowledge of its distribution and abundance at low latitudes, where it plays the dominant role in atmospheric chemistry and thus the production of the hydroxyl radical (OH), the primary oxidizing agent in the troposphere. It is also in this region where accurate measurements are most sparse. Additionally, because of its relatively short lifetime, accurate extrapolations from measurements at surface stations cannot be used to determine how much ozone is present over vast oceanic expanses. For these reasons, it appears that space-based remote sensing provides a critical method by which the distribution of tropospheric ozone can be determined.

II Satellite tropospheric ozone measurements on timescales of months to years

Satellite measurements of ozone have been made since the launch of the Backscatter Ultraviolet (BUV) instrument on the US Nimbus-4 satellite in 1970 (Heath *et al.*, 1975). Subsequent instruments, using the same measurement technique as the Nimbus-4 BUV instrument, have been providing total ozone measurements almost continuously since that time. These instruments provide quantitative information about the total amount of ozone between the Earth's surface and the satellite and, primarily, provide scientists with information about the distribution of ozone in the stratosphere, since approximately 90% of the ozone in the atmosphere lies in this region (approximately 15–55 km). The remaining 10% is located in the troposphere; the boundary between the troposphere and the stratosphere, the tropopause, is generally located at an altitude of between 12 and 18 km and varies as both a function of latitude and time of year. In the tropics, the tropopause is located higher in the atmosphere (16–18 km) than at middle

latitudes, where the height of the tropopause can be as low as 8–10 km, or as high as 15–16 km, with higher tropopause heights generally found in the summer.

The amount of ozone in a column of air is expressed in units called Dobson Units (DU) where 1 DU has a value of 2.69×10^{16} molecules of ozone/cm². A representative amount of total ozone in the atmosphere is 300 DU, of which approximately 30 DU is in the troposphere and the remainder is in the stratosphere. In the unusual occurrence of the 'ozone hole' found over Antarctica during austral spring, total ozone values <100 DU have been measured.

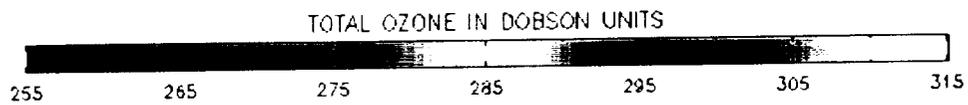
In a series of papers that date back to the mid-1980s, our research group has pursued the idea that satellite data sets can be used to glean otherwise unattainable insight into the distribution of ozone in the troposphere. Although the Total Ozone Mapping Spectrometer (TOMS) was built to monitor ozone in the stratosphere, and perhaps provide insights into stratospheric ozone depletion, Fishman *et al.* (1986) showed that enhancements in the total ozone signal over Brazil occurred when widespread biomass burning was present. From the monthly average of the TOMS measurements shown in Figure 1a, enhanced values can be seen over South America and southern Africa and in a region over the South Atlantic Ocean, in what appears to be a plume coming off the African continent during October 1987. During other months, such as October 1985 (Figure 1b), total ozone enhancements can be found off the west coast of North America. During this particular month, a persistent area of anticyclonic circulation was situated off the northwest coast of the United States, resulting in the plume-like structure emanating from the highly populated Los Angeles–San Diego region.

The data depicted in these figures, however, reflect total ozone measurements, of which only a relatively small percentage is found in the troposphere. At higher latitudes, meteorological activity is generally more vigorous and persistent patterns over the period of a month are rare. Thus, identification of enhanced ozone of tropospheric origin is difficult and quasi-persistent plumes from Europe and northern Asia would be lost beneath the variable stratospheric ozone amounts that would overwhelm any persistent tropospheric enhancements in these regions. Thus, persistent ozone sources in the tropics offered the first indications that satellite information could be used to identify ozone pollution sources.

Through the 1990s, our research group has focused on trying to extract information about the troposphere from satellite measurements by assuming that ozone variability in the stratosphere is defined on relatively large spatial scales compared with the troposphere, and that information could be obtained about the troposphere if this larger scale stratospheric component could be isolated. Once the stratospheric ozone distribution has been established, we can then examine the 'residual' information contained in the TOMS total ozone measurements to infer information about the troposphere. Thus, the fundamental presupposition of the residual technique is that useful information about tropospheric ozone could be inferred from satellite measurements if the amount of ozone in the stratosphere could be quantified and then subtracted from the total ozone derived from TOMS data. The resultant distribution derived from this method was referred to as the 'tropospheric ozone residual' (TOR). The stratospheric ozone distribution that was subtracted from the concurrent TOMS total ozone measurement was first derived using the Stratospheric Aerosol and Gas Experiment (SAGE) data. SAGE has been providing ozone information since 1979 when it was first launched on the Atmospheric Explorer Mission 2 (AEM-2) satellite, which functioned for nearly three

years. In 1984, SAGE II was launched on the Earth Radiation Budget Satellite (ERBS) and has provided measurement for more than 15 years. Although still providing measurements in the year 2000, degradation of the orbit and some of the instrument components have resulted in less useful measurements being obtained 16 years after launch than had been provided earlier.

(a)



(b)

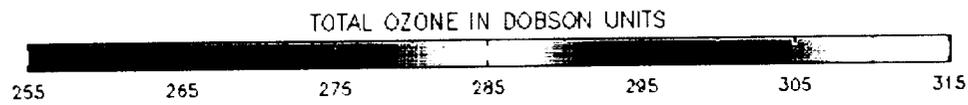
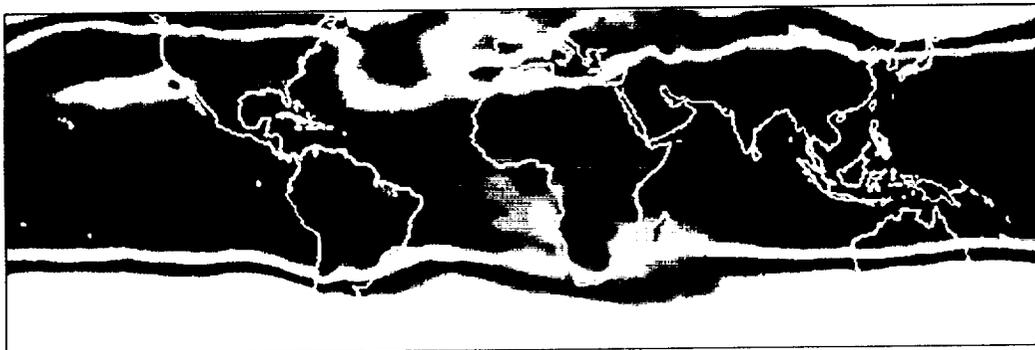


Figure 1 Monthly average values of TOMS total ozone for: a) October 1987; b) October 1985

Using standard tropopause height information from the National Center for Environmental Prediction (NCEP) in conjunction with the SAGE profiles, Fishman *et al.* (1990) demonstrated that a fairly accurate climatological depiction of tropospheric ozone could be obtained to include a domain that included both the tropics and middle latitudes (approximately 50°N to approximately 50°S). The seasonal climatology of the TOR is depicted in Figure 2 (Fishman *et al.*, 1990) and shows the elevated amounts of tropospheric ozone present in the middle latitudes during northern hemisphere summer; elevated TOR values are also present in the tropics and subtropics in the southern hemisphere during austral spring (September–November), a consequence of widespread biomass burning during that time of the year.

The TOR technique using SAGE data has generally been limited to climatological studies; a more rigorous analysis of the data indicates that the TOMS/SAGE TOR is not useful to study tropospheric ozone variations at nonclimatological scales because the TOMS/SAGE TOR is subject to sampling aliasing for periods of seasons or less, a consequence of the approximately 40-day period needed to obtain complete latitudinal coverage (Vukovich *et al.*, 1996). On the other hand, a variation of the TOR methodology, called the convective cloud differential (CCD) technique, has been used to show interannual variability of tropospheric ozone in the tropics (Ziemke *et al.*, 2000). The CCD technique uses information in the TOMS measurements taking advantage of the fact that TOMS does not see below cloud tops. In the tropics, Ziemke *et al.* (1998) assume that highly reflective clouds reach to the tropopause and, thus, a measure of the amount of ozone throughout the stratosphere (and above) can be obtained when the reflectivity is above 0.9. Ziemke *et al.* then seek out nearby regions where no clouds are present and assume that the total ozone at these locations is truly representative of the amount of ozone in both the stratosphere and troposphere. They then subtract these two quantities to derive the amount of ozone in the troposphere (or tropospheric column ozone, TCO), a quantity analogous to the TOR described by Fishman *et al.* (1990).

The tropical TCO distributions from three consecutive years 1996–98 are shown in Figure 3. The middle depiction illustrates the impact of unusually high amounts of biomass burning in southeast Asia on the resultant TCO distribution. Although biomass burning in this region of the world is a common practice, the amount of burning was exceptionally great during the strong El Niño of 1997–98 because of the extremely dry conditions in that region of the world. Satellite data have also found enhanced pollution over this region in 1983 and 1994 (Fishman *et al.*, 1990; Reichle *et al.*, 1999; Rinsland *et al.*, 1999).

III Daily TOR maps for studies of long-range transport

To achieve better temporal resolution, concurrent measurements from TOMS and Solar Backscatter Ultraviolet (SBUV) instruments have been used to derive daily TOR distributions. The procedure to derive the TOMS/SBUV TOR also uses knowledge of tropopause height obtained from gridded fields determined from the NCEP assimilation/modeling data base. The grid that is used for the calculations is a 100 by 288 matrix, having a resolution of 1° latitude by 1.25° longitude and covering the region between the latitudes of 50°S and 50°N. In the region 50°S to 50°N, the SBUV

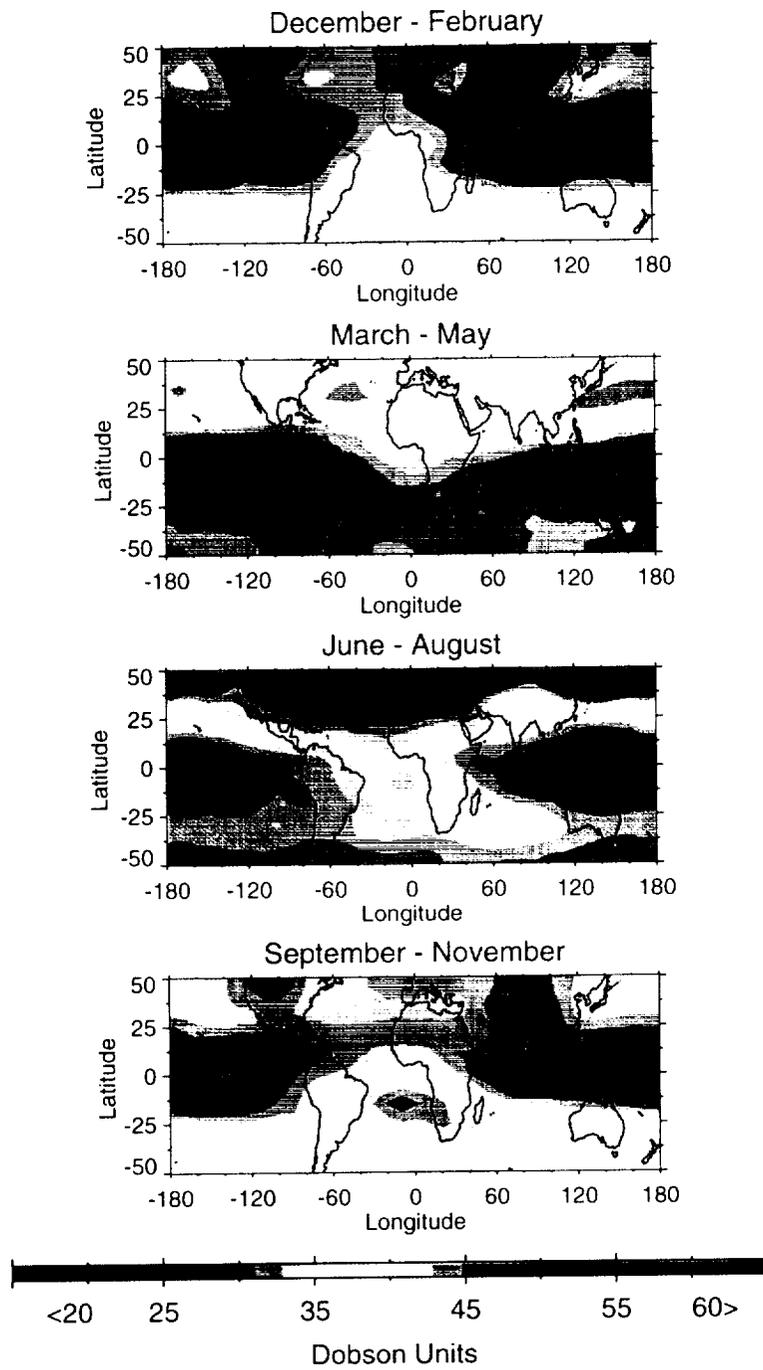


Figure 2 Seasonal climatology of tropospheric ozone derived from the TOMS and SAGE data sets using the tropospheric ozone residual method
Source: Fishman et al. (1990)

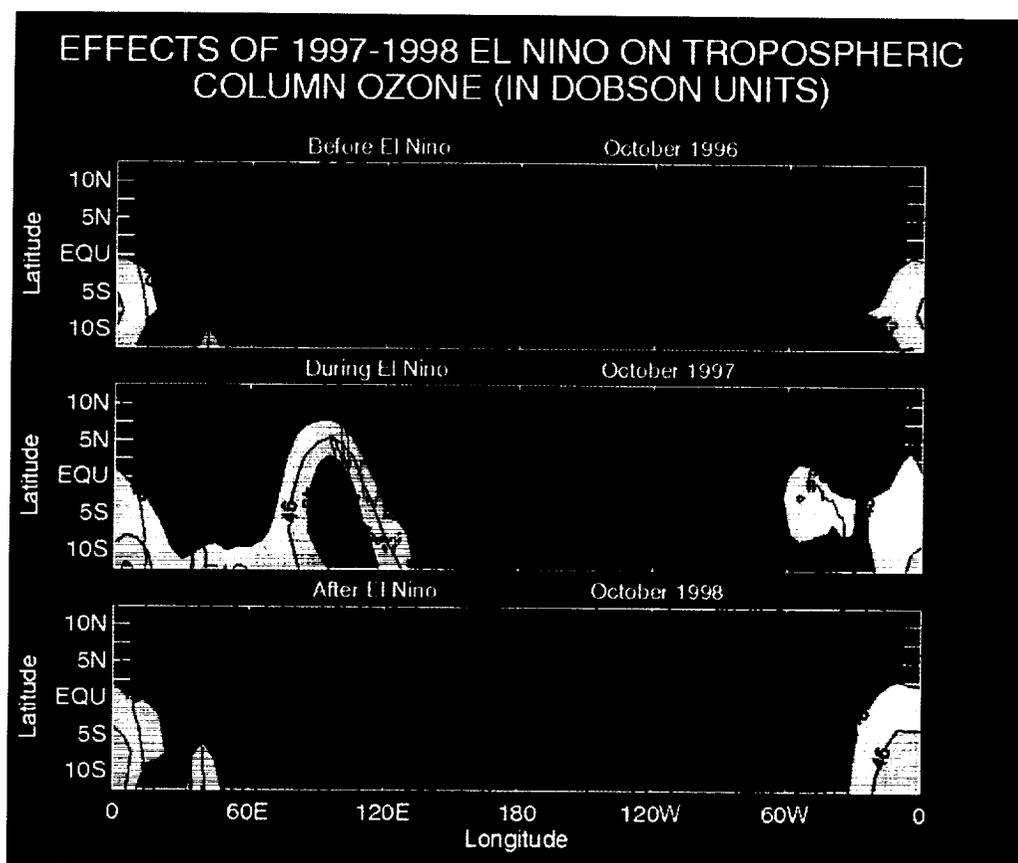


Figure 3 Tropospheric ozone derived from the tropical tropospheric ozone column method described in Ziemke *et al.* (2000). The three panels depict the interannual variability for October 1996, 1997, 1998, and illustrate the enhancement in 1997 owing to widespread biomass burning because of the dry conditions in southeast Asia resulting from the strong El Niño circulation present during that year

instrument provides approximately 750–800 vertical profiles on a daily basis (SAGE, on the other hand, provides about 30 vertical profiles per day). After the stratospheric ozone is determined at each grid point where SBUV data are available, an interpolation procedure is used to fill in all missing values within the daily matrix. A 5-day running average is applied using the daily matrices of the SBUV stratospheric ozone and the resulting matrix is used to represent the stratospheric ozone distribution for the central day (i.e., day 3) used in the 5-day running average. The TOMS data, on the other hand, are not temporally averaged. The global TOMS/SBUV TOR distribution is determined for a given day by subtracting the 5-day running averaged SBUV stratospheric ozone data from the TOMS total ozone data at each grid point.

Using this technique, the daily TOR were compared with data obtained from the TRACE-A field mission in 1992 (Fishman *et al.*, 1996b). In general, except when heavy

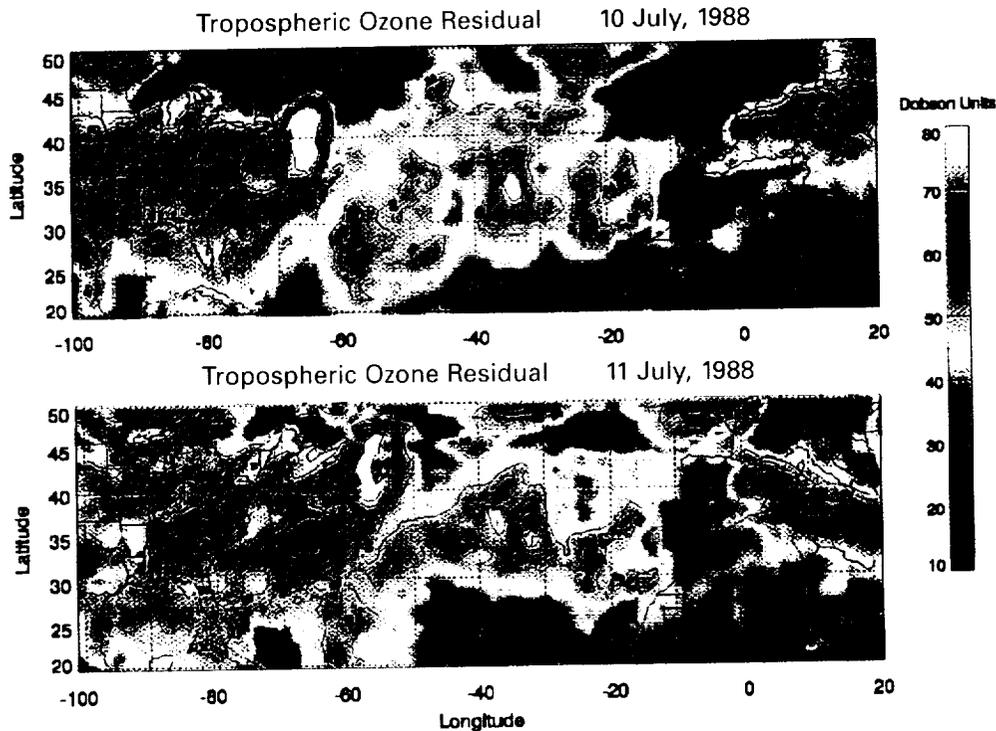


Figure 5 Two panels showing consecutive days of the distribution of tropospheric ozone across the northern Atlantic Ocean. The satellite analysis employs a modified version of the technique described by Fishman and Balok (1999); see text for discussion

persistent meteorological situation that resulted in the formation of the air pollution episode over the eastern USA between 4 July and 10 July started to change on the 9th and that the high amounts of ozone off the east coast on the 10th were transported relatively rapidly to the east. The depictions for 10 and 11 July (Figure 5a, upper and lower panel, respectively) show that the high amount of TOR centered at 40°N , 66°W on the 10th has moved approximately 1000 km east-northeast on the 11th.

One of the forthcoming issues to emerge recently is the impact of intercontinental transport on regional air quality. Certainly the best way to gain insight into this problem is through the use of satellite measurements, and the data shown in Figure 5 provide an example of how pollution over North America is transported over considerable distances; or as shown in Figure 4, how emissions can be recirculated under the right meteorological conditions resulting in the formation of even higher amounts of ozone pollution. The question we currently address is, 'What eventually happens to the ozone pollution observed in this US episode, and does any of it reach Europe?'. The trajectory calculations shown in Figure 6 provide some insight into that question.

The four panels in Figure 6 summarize a set of 6-day forward trajectories that start at two points (41°N , 67°W ; and 39°N , 67°W) located within the maximum situated off the east coast of the USA on 10 July. Each of the four panels represents calculations for different starting altitudes within the troposphere: 1500 m; 3000 m; 5000 m; and 8000 m.

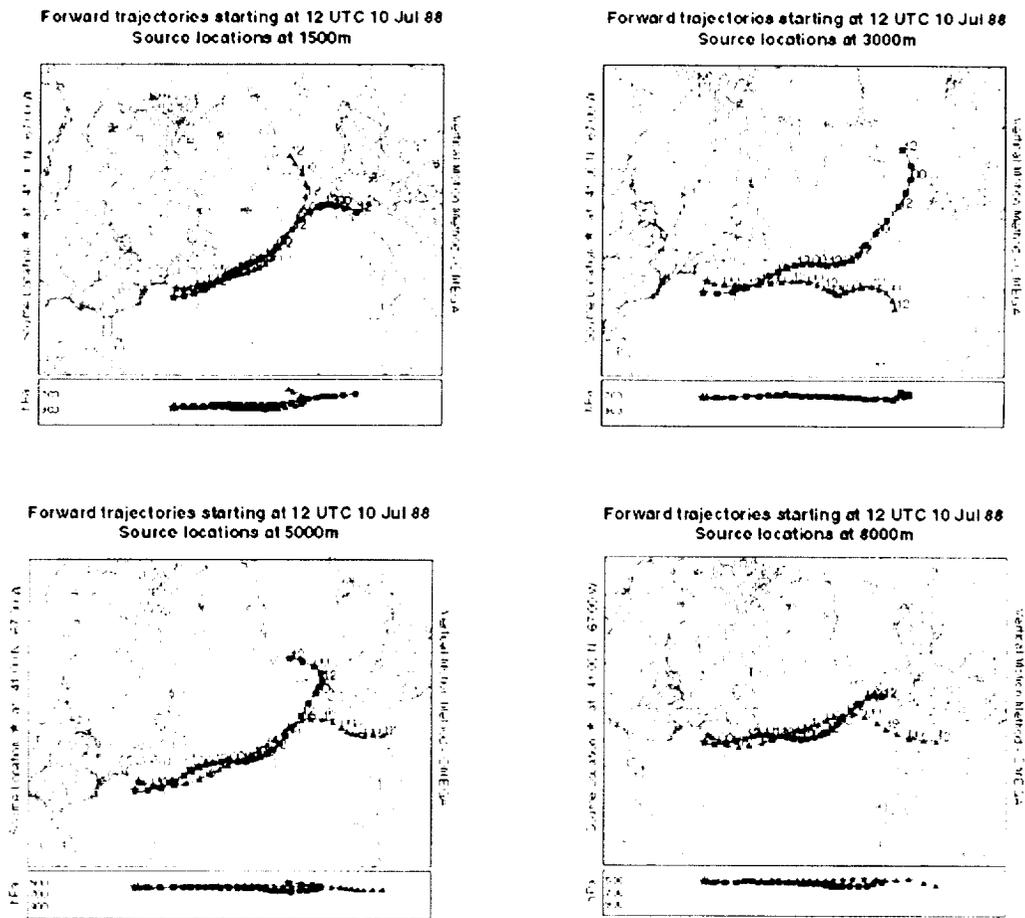


Figure 6 Forward trajectories originating at 39°N , 67°W , and 41°N , 67°W for 10 July, 1988. Both calculations are for 6 days and the symbols show air parcel locations every 6 hours. The lower portion of the different panels shows the altitudes at which the trajectories are located. Within each panel, the red triangles show the position of the air parcel that originated from the northern initial starting point (red star), whereas the blue squares represent the locations on the air parcels at the southern starting point (blue star)

Since the TOR is an integral of all the ozone within the tropopause, it is important to note that vertical wind shear incorporates great uncertainty into the interpretation of where parcels of air may be transported. Of the eight trajectories shown here, only two or three of them suggest that the elevated pollution levels from North America are located over Europe during the next six days. Of perhaps even greater significance is the fact that the air is undergoing chemical reactions along these trajectories, and it is likely that much of the ozone observed by satellite off the east coast of the USA may have been depleted as it tracks eastward. It is also noteworthy that points originating only approximately 200 km apart initially can diverge by several thousand kilometres

aerosols were present owing to widespread biomass burning, the TOR generally captured the spatial gradient of tropospheric ozone, but usually underestimated the magnitude of the gradient, and nominally captured only approximately 60% of the spatial difference between two points when compared with gradients determined from aircraft measurements (Fishman *et al.*, 1996a). Further analysis by Fishman and Balok (1999) found that the climatological distribution of the archived SBUV data was critically inaccurate in the lowest three levels of the atmosphere. In turn, these inaccurate values were the primary contributing factor to the poor agreement found between the SBUV/TOMS TOR distributions and the SAGE/TOMS TOR noted by Vukovich *et al.* (1997). After analysing thousands of soundings from more than 20 ozonesonde stations, Fishman and Balok employed an empirical correction to the archived SBUV measurements to derive stratospheric integrals that could be subtracted from the concurrent TOMS observations. These data were then used to characterize the development of an air pollution episode over the eastern USA.

Figure 4 schematically summarizes the findings in Fishman and Balok (1999) that showed that a pollution episode over the northern USA may have been the precursor for the establishment of a widespread episode that developed over southern USA three and four days later. The two panels on the left show color depictions of surface ozone on two different days: 6 July 1988 (top) and 9 July 1988 (bottom). The high surface concentrations are depicted by the areas of red.

Because of the intense anticyclone that persisted over much of the eastern USA during this time, widespread stagnation developed and pollution built up behind the stationary front over the western Atlantic Ocean (upper right panel). Note that the high pollution seen in the upper left panel for 6 July had begun to build up on 4 July (not shown). Thus, the pool of pollution seen off the coast of the Carolinas on 6 July determined from the satellite data had formed from emissions and subsequent photochemical generation for several days previous. The lower right panel shows that pollution from the red area (TOR <75 DU) is then carried to the southern states and elevated ozone is seen at the surface several days later (9 July). To derive the TOR shown in this figure, the SBUV data were empirically corrected using the climatology derived from the ozonesondes from Wallops Island (37°N, 75°W), since this is the only site available within this entire region. To expand this type of methodology to larger regions, data must also be used from other sites.

A modification to the empirical correction described in Fishman and Balok (1999) was recently achieved through the use of the seasonally dependent three-dimensional data set developed by Logan (1999). Thus, the newest methodology for computing TOR fields incorporates an empirically corrected value where the corrections are a function of latitude and longitude, as well as month. With this empirical correction, all SBUV values have the ratio between the amount of ozone in the lowest three layers that had been determined from the Logan climatology. This methodology is then applied to the case study described below to examine whether or not the ozone that had built up over the USA described in Fishman and Balok is transported to Europe.

Figure 5 shows the TOR distribution determined from the above methodology for 10 and 11 July 1988, the end of the pollution episode discussed in Fishman and Balok (1999). The longitudinal domain has been greatly expanded and now uses information for the empirical correction from some European ozonesonde stations, in addition to the measurements at Wallops Island. Fishman and Balok (1999) showed that the

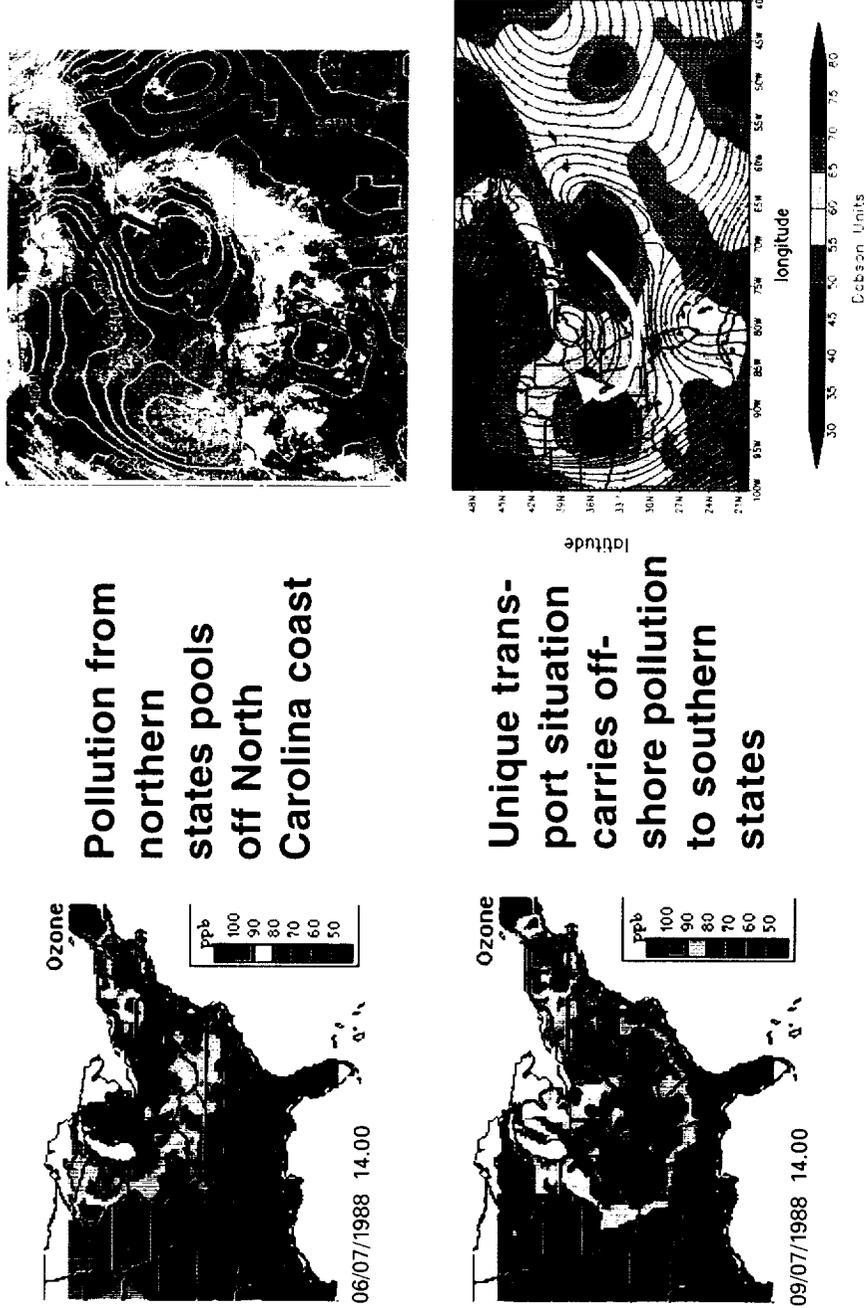
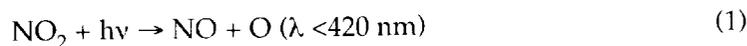


Figure 4 A four-panel collage summarizing the Fishman and Balok (1999) study showing how pollution originating over the northern USA eventually was transported to the southern part of the country several days later. The satellite depictions in the right panels depict how tropospheric ozone pooled off the east coast of North Carolina during the episode, serving as a reservoir for subsequent transport that led to one of the worst pollution episodes over the southern USA
 Source: Fishman and Balok (1999)

after 6 days. One must also keep in mind that there are limitations to the use of such trajectory models, especially after running such calculations for more than 2–3 days; nonetheless, these depictions illustrate the complexity of atmospheric motion.

IV Measurement of other trace gases and future capabilities

Unlike most other pollutants found in the troposphere, ozone is not emitted directly but, instead, is a product of photochemical reactions. Photodissociation of NO_2 by (visible) sunlight is the only significant anthropogenic source of O_3 in the troposphere



immediately followed by



where the M in reaction (2) represents any nonreactive molecule that absorbs some of the excess energy of the intermediate product formed in the reaction (2).

The atmospheric oxidation of CO is initiated by reaction with the hydroxyl radical (OH):



The hydrogen atom combines with an oxygen molecule in a three-body reaction:



to form the hydroperoxy radical, HO_2 . The peroxy radicals (HO_2 and longer chain analogs, RO_2 , where R represents a methyl, or other nonmethane hydrocarbon radical) are key for converting NO to NO_2 :



so that OH is recycled and remains in the atmosphere to initiate new oxidation sequences. The net reaction for this particular example is:

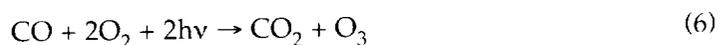


Figure 7 depicts information about the distribution of tropospheric nitrogen dioxide (NO_2) derived from a technique analogous to the TOR methodology that assumes that tropospheric information can be derived from total column measurements of NO_2 . These measurements have been obtained from the Global Ozone Measurement Experiment (GOME) instrument on the ER-1 Satellite launched in 1995. A similar instrument, but with advanced capability, called SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Cartography) will be launched on the ESA Envisat satellite in 2001 and should be able to produce similar data with better spatial and temporal resolution (see Burrows, 1999, for additional details).

An instrument to measure CO, called MOPITT (Measurement of Pollution in the Troposphere) was launched in December 1999 on the USA's Earth Observing System Terra satellite, and is providing the first measurements of CO

Average Tropospheric NO₂ Column Density During 1997, GOME

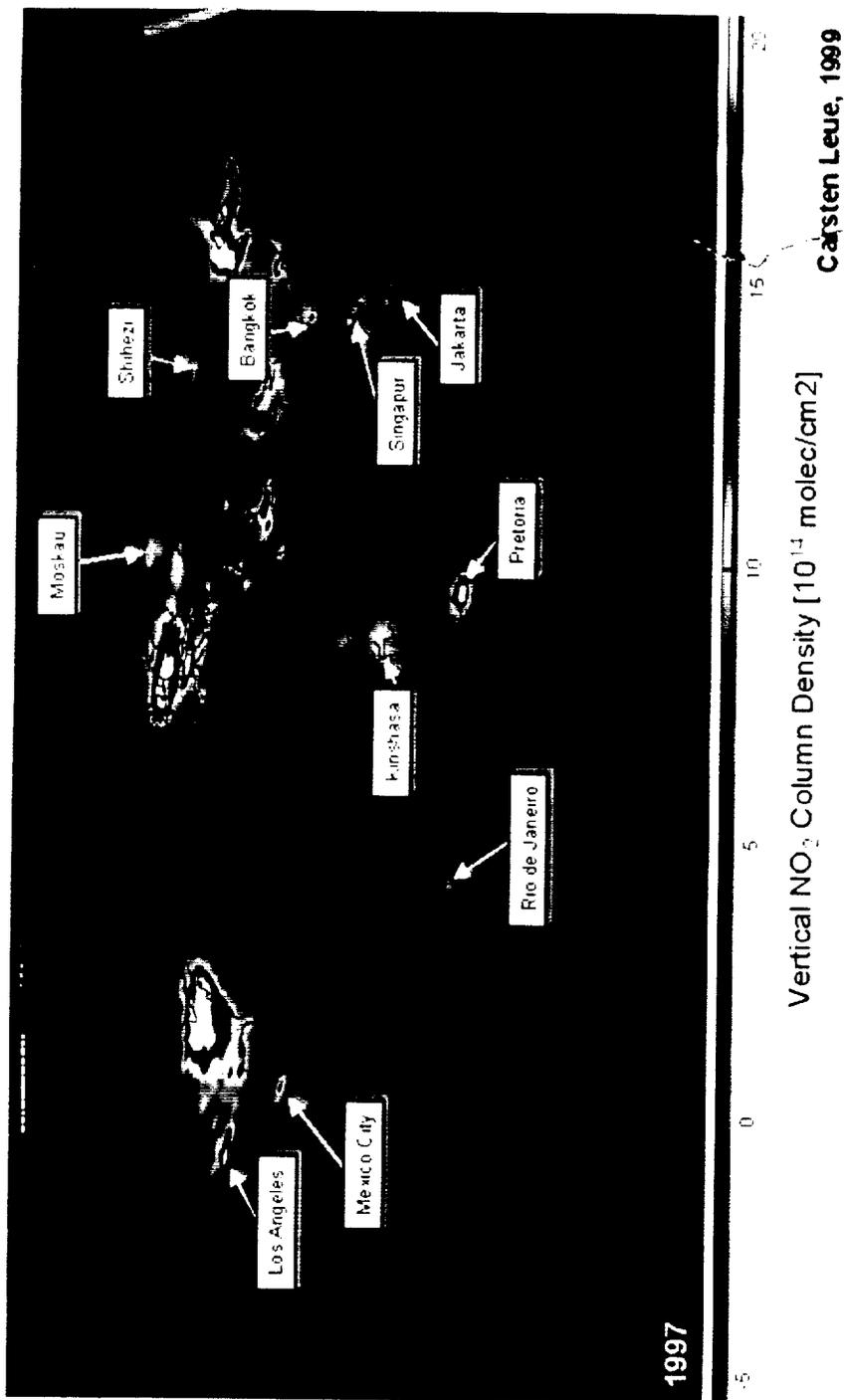


Figure 7 Excess column NO₂ (from which the tropospheric component is inferred) distribution derived from GOME data
Source: Industrial pollution TEM mean NO₂ column density from GOME 1997 A. Richter and J.P. Burrows, personal communication (2000)

(information about and data from MOPITT are available electronically at <http://www.eos-am.gsfc.nasa.gov/mopitt.html>). Thus, when concurrent CO, NO₂, and O₃ measurements become available, our understanding of the processes that result in the formation of tropospheric ozone will be enhanced immeasurably. Direct measurements of tropospheric ozone will be available from the Tropospheric Emissions Spectrometer (TES), which is scheduled to be launched on the US Earth Observing System Aura platform in 2003. This measurement capability will also greatly enhance our knowledge of tropospheric ozone.

In summary, the measurement of tropospheric ozone from space has been demonstrated using indirect methods from satellites whose technology was developed in the 1970s. As the new millennium begins, the satellite community has also demonstrated the capability to measure important precursors to *in situ* tropospheric ozone production and these measurements will improve significantly before the end of the decade. By that time, we should be able to gain fundamental understanding into not only the distribution and variability of tropospheric ozone, but also into the transport and chemical processes that result in its formation.

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