GLOBAL TRANSPORT OF OZONE

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ABSTRACT: The three principal mechanisms for large scale atmospheric transport of tropospheric ozone [injection from the stratosphere, transport from regions of net production in the boundary layer and distribution of O_3 precursors resulting from either stratospheric injection or surface emissions] are examined in the light of current observations. While the actual O_3 climatology may be much more complex than it currently appears, the limited data suggests that ozone in the Southern Hemisphere and the northern tropics and subtropics is strongly influenced by transport from the stratosphere. At this time, the major questions are in the southern tropics and the northern mid-latitudes. The high levels of ozone observed over South America appear to be either the result of local chemical production or transport from higher latitudes. Both the latitude gradient and the seasonal cycle in the northern mid-latitudes suggest a significant, if not dominant, role for the transport of both O_3 and its precursors from source regions in the boundary layer, though transport from the upper troposphere also pays a role.

1. INTRODUCTION

Recent observations suggesting long term increases in CO and CH_4 (e.g. Rasmussen and Khalil, 1984; Rinsland and Levine, 1985) have raised questions about the long term levels of tropospheric ozone. Before speculating on trends, the chemical and meteorological processes that control the current level of O_3 must be understood. We have known for many years that tropospheric ozone is both transported from the stratosphere and produced photochemically in the polluted boundary layer. In 1973 a photochemical theory was proposed for O_3 in the general troposphere (Crutzen, 1974; Chameides and Walker, 1973). The relative importance on a global scale of net photochemical production and direct stratospheric injection are still in question, though recent estimates of global photochemical production and loss exceed the estimates of stratospheric injection (e.g. Liu, this volume). Recently, net photochemical production of ozone in the upper

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troposphere driven by NO_x injected from the stratosphere has been proposed (Liu et al., 1980), and large net production in the more remote boundary layer has also been suggested (Liu, this volume).

After a brief discussion of the basic mechanisms of large scale transport and chemical production, we will consider the existing observations. Two regions in particular, the tropics and the northern mid-latitudes raise questions that are not easily explained by either local chemical production or large scale transport processes. Some future studies will then be proposed.

BASIC PROCESSES

2.1 Continuity Equation

The time dependent behavior of the ozone mixing ratio, R, is described by the following continuity equation,

$$\partial R/\partial t = - \mathbf{V}_3 \nabla_3 R + P - LR - DR \qquad (1)$$

where P is the rate of chemical production, L is the rate coefficient for chemical destruction and D is the rate coefficient for surface destruction. The first term represents the 3-dimensional advection of R by the winds, the next two comprise the chemical tendency and the last one is surface destruction. Note that this equation is written for the mixing ratio of ozone, not the number density. The motions of the fluid parcels do not depend on the ozone concentration and, assuming no production or destruction, conserve the global ozone mixing ratio. Transport, in an Eulerian sense, depends on the existence of a gradient either horizontal or vertical, in the ozone mixing ratio. The rates of chemical production and destruction, while a direct function of number density, are easily converted to mixing ratio.

2.2 Transport

The largest gradients in the mixing ratio of O_3 are found between the stratosphere and the troposphere where the values drop from 7-10 ppm at 10mb to .06-.01 ppm at the ground. A simple description of the downward transport of O_3 from the stratosphere is provided by a diabatic circulation with rising motion in the tropics and sinking motion at higher latitudes, the same mechanism that carries heat poleward and maintains the latitudinal structure of the tropopause. The downward branch reaches a maximum at mid-latitudes and is strongest in the winter and spring. A detailed description is given by Mahlman et al. (1984). Growing extratropical cyclones tap the O_3 accumulating in the lower stratosphere and transport it into the troposphere. This process has been described as a "tropopause fold event" (Danielsen, 1968). These events carry the O_3 downward and equatorward along an anti-cyclonic path and are most prevalent at mid-latitudes in the spring. As a result, the stratospheric injection of O_3, with a global

average flux in the range 3-12 x 10^{10} molecules $cm^{-2}sec^{-1}$, shows a maximum at mid-latitudes in the spring.

The transport of O_3 from the boundary layer is much more difficult to estimate. While the mean flow is toward the east(westerly) in the mid-latitudes and toward the west(easterly) in the tropics, the actual day to day winds are highly variable. Given both significant destruction and weak winds at the surface, effective long-range transport requires that the boundary layer air be lifted into the "free troposphere." This can be accomplished by processes ranging from a single convective cloud to a large synoptic-scale storm. However, the development of high O_3 levels in the boundary layer is normally associated with stable conditions implying limited vertical transport. An alternative is the transport of precursors for O_3 formation, rather than O_3 itself. The resulting O_3 formation becomes a product of both transport and chemistry.

2.3 Chemistry

While the magnitude of the O_3 chemical tendency in the clean "free troposphere", which ranges from 0.1-0.2 ppb/day in the winter to ~5.0 ppb/day in the summer, depends on many chemical species and physical parameters, the sign of the tendency depends most strongly on NO_x (Levy et al., 1985). When many non-methane hydrocarbons are available, the chemistry becomes much more complex, but the level of O_3 still remains most sensitive to the level of NO_x (e.g. Liu et al., 1987).

Since the vertical lifting is generally accompanied by condensation, the large scale transport of the key precursor, NO_x, becomes quite complicated. One of the reaction products, HNO_3, is very soluble, while another, PAN, is not. Thus, the indirect transport of O_3 via its precursors from source regions in the boundary layer requires an understanding of the transport and chemistry of the reactive nitrogen species.

3. OBSERVATIONS/GLOBAL TRANSPORT

Before attempting to assess the contributions of global transport to the climatology of tropospheric O_3, we will briefly examine the available observations. While there have been a number of measurement programs, some with time series of 10 years or more, the distribution of vertical profiles is highly skewed toward the populated northern mid-latitudes with only 3-4 stations located in the Southern Hemisphere (S.H.).

3.1 Global Tropospheric Climatology

To briefly summarize the detailed analysis of this limited data set by Logan (1985): O_3 increases poleward from the tropics and levels off at mid-latitudes where it shows a summer maximum in the Northern Hemisphere (N.H.); averaged over the year, O_3 appears to be more

abundant in the N.H. than in the S.H. though the data is very limited; O_3 generally increases with height, except in the severely polluted boundary layer; throughout the "free troposphere," excluding continental northern mid-latitudes, the seasonal maximum occurs in that hemisphere's spring and the seasonal minimum in the fall or winter; the seasonal cycles in the boundary layer show no simple pattern and depend on the local sources and sinks. This relatively simple picture of the O_3 climatology may be due in large part to the sparse and highly skewed data sets.

Currently, there are two major exceptions: the large values found in the lower and mid-troposphere near South America in the southern tropics during October (Logan and Kirchhoff, 1986); the extension of the spring maximum into the summer up through 500 mb for continental mid-latitudes in the N.H. (London, private communication; Logan, 1985).

3.2 Stratospheric Injection

Excluding the two anomalies, the global climatology of tropospheric O_3 is quite consistent with stratospheric injection followed by equatorward/downward transport in the troposphere. The observed latitude gradients, interhemispheric differences, vertical gradients and seasonal cycles are all qualitatively consistent with a recent simulation of this process by a medium resolution general circulation/transport model, though model defects are believed to exaggerate the interhemispheric gradient and to produce excessive levels of O_3 at northern high latitudes (Levy et al., 1985).

A more global data base may find a much weaker interhemispheric gradient. While stationary eddies and planetary wave activity are much weaker in the S.H., transient eddies are both more frequent and stronger. Therefore, both hemispheres may do an equivalent job of tapping their respective lower stratospheres for O_3. Any hemispheric difference would then be the result of differences in that hemisphere's diabatic circulation.

3.3 Tropical "Anomalies"

Until recently, the few available measurements of tropical O_3 resulted in a simple picture (small seasonal variability, low values, slight increase with height) that was consistent with stratospheric injection and the strong vertical mixing expected in the tropics from convection. However, Kirchhoff et al., (1984) and Gregory et al., (1984) found very high values over South America during October and further analysis by Logan and Kirchhoff (1986) found a strong September - October maximum over Natal, Brazil (6°S, 35°W) all the way up through 500 mb. These observations raise two questions: What is the source of the high O_3? What is the spatial and temporal extent of the maximum?

Dry season burning, which has been associated with an increase in the boundary layer concentration of both O_3 and O_3 precursors (Crutzen et al., 1985), has been proposed as a source of the observed maximum (Logan and Kirchhoff, 1986). How this produces a maximum off both

coasts of South America is not clear, but the observed increase over Natal does grow during the burning season (Logan and Kirchhoff, 1986). Furthermore, Marenco (this volume) reports high O_3 throughout the troposphere over Africa in association with burning and Fishman (this volume) infers high values over a region running from western South America to eastern Africa during periods of extensive biomass burning. However, while some individual profiles over Natal may show a maximum at 700 mb, the monthly means increase with height up to 500 mb, and the largest excursions from "normal" tropical O_3 values appear to be highly correlated with sharp drops in relative humidity. These last two observations point to the excess O_3 being transported from above, and recent simulations with a high resolution GCM indicate equatorward transport from 60°S by the strong transient eddies (Mahlman, private communication).

Whether the seasonal maxima result from O_3 transported down from the upper troposphere at high latitudes or from O_3 and O_3 precursors transported from regions of biomass burning at the surface, large scale transport plays a major role. If the former, the maxima should be widely distributed and not necessarily so sharply seasonally dependent. If the latter, the seasonal maxima should be highly correlated with biomass burning and other products of combustion should be present.

3.4 Mid-latitude Summer Maxima

While the analysis of Logan (1985) shows a spring maximum at 300 mb for the multi-year average of continental ozonesondes in the N.H., at 500 mb and below this becomes a broad spring-summer maximum for continental stations at mid-latitudes. Recent studies of early O_3 measurements (Kley, this volume; Logan, 1985; Bojkov, this volume) find that the 1880 measurements in Paris were significantly lower than present values, that the seasonal maximum at Arosa previously occurred in the spring, and that a number of mid-latitude stations show increases over the last 10-20 years. It would seem that there has been an increase in tropospheric O_3 at mid-latitudes during the 1900's and that this increase has been most apparent in the summer. The coincidence of a rapid increase in the emission of O_3 precursors during this period and the largest observed increase in O_3 occurring during the summer, a time of maximum photochemistry, is hard to ignore. One should note, however, that the seasonal pattern at a station frequently varies with height in a given year and from year to year at a given height (Levy, 1985). This suggests that transport of the O_3 precursors is important.

4. PROPOSED STUDIES

4.1 Global

There is a great need for sufficient O_3 data to create a realistic global climatology, particularly in the S.H. where the only O_3 profile

measurements are at Samoa, Aspendale, Natal and Antarctica. While the satellite studies reported by Fishman in this volume may point out regions where there is O_3 variability, the technique is limited to the low latitudes and is not sufficiently quantitative. A program to measure O_3 profiles in the tropics and over both oceans of the Northern Hemisphere is needed.

4.2 Tropics

It is clear that this region is far more complex than was thought previously. It is particularly important to determine the spatial and temporal extent of the O_3 maxima and their relation to the locations and times of biomass burning. We not only need more measurements of O_3, but we need to include measurements of O_3 precursors such as CO and NO_y and chemical tracers of the air parcel's past history.

4.3 Northern Mid-latitudes

Extensive O_3 measurements are needed to determine whether the yearly increase and the summer maximum are local, regional or hemispheric phenomena. Both surface and vertical profile data over the Atlantic and the Pacific would help. Measurements of reactive precursors as well as chemical tracers of atmospheric transport should be included with the O_3 observations.

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