

# Ozone, Aerosols, and Solar Radiation in the Stratosphere

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The Climatic Impact Assessment Program (CIAP) of the Department of Transportation is—among other things—faced with the task of providing a detailed review of the “undisturbed” stratosphere as it exists before the commencement of SST traffic. CIAP Monograph No. 1, presently in preparation, deals with this “natural stratosphere.” Material for a rough draft of this volume was compiled by approximately 90 scientists during a workshop in Ft. Lauderdale from November 28–December 1, 1972. This short review paper, based upon the draft of CIAP Monograph No. 1, points out the highlights of our present knowledge of the stratosphere, especially with respect to the  $O_3$ ,  $NO_x$ ,  $H_2O$  problems, and also explores some of the deficiencies in this knowledge which will have to be overcome before a complete assessment of human impact on this atmospheric region can be made.

## The Ozone Problem

OZONE, even though a minor constituent of the atmosphere ( $4 \times 10^{-7}$  parts by volume average concentration), derives its eminent importance from its capability of blocking UV radiation at wavelengths  $<3000 \text{ \AA}$ . Dobson instruments (ca. 100 observation points globally) provide the major data source on total ozone. Detailed vertical structures have been detected from infrequent and intermittent balloon measurements. Rocket and aircraft data also are listed in the literature. Satellite data (Nimbus III and IV) carry great promise for global and detailed observations of total ozone. However insufficient funds and manpower are provided to analyze even the presently available data bank.

## Latitudinal Distribution

Lowest values during spring are observed at  $5^\circ$  to  $10^\circ\text{S}$ , and on an annual average close to the equator, but slightly south of the “meteorological equator.” In the northern hemisphere highest total ozone values appear during spring in polar latitudes, in the southern hemisphere in middle latitudes (Fig. 1). This is conclusive evidence that the photochemistry of ozone is strongly overshadowed by transport processes. If photochemistry alone were involved, both hemispheres should show the same seasonal behavior.

## Seasonal Variation

A maximum of total ozone is found in both hemispheres in middle latitudes during spring. Polar latitudes in the northern hemisphere also reveal a spring maximum of total ozone (Fig. 1) indicating that horizontal and vertical large-scale eddy transport processes in the lower stratosphere are supplying middle and high latitudes in the northern hemisphere with  $O_3$  at a very efficient rate. In the southern hemisphere the eddy transport processes between middle and polar latitudes proceed slower, causing the time lag with which the ozone maximum is observed over the south pole.

Cyclone activity causes most of the ozone transport—vertically as well as horizontally—in middle latitudes. This can be shown from “ozone waves” that can be detected by satellites (Fig. 2). These waves exhibit the same

wavelengths, phase- and group-velocity characteristics as traveling cyclonic disturbances. A well developed planetary wave number 2 in the northern hemisphere stratosphere must be held responsible for the efficient transport of  $O_3$  into north-polar regions during spring. In the southern hemisphere only planetary wave number 1 (eccentricity of the polar vortex) is reasonably well developed. The higher wave numbers 2 and 3 are rather insignificant there. This causes the  $O_3$  transport by standing planetary waves in the southern hemisphere to be very much less significant than in the northern hemisphere. Lower transport efficiency of pollutants generated in the lower stratosphere (near 20 km) should be expected, therefore, in the south polar regions than in the north polar regions.

At this time we have only very sketchy information about the meridional extent of the aerosol layer near 15–20 km to polar regions. Some evidence exists that the aerosol layer is present near about 15 km over Alaska

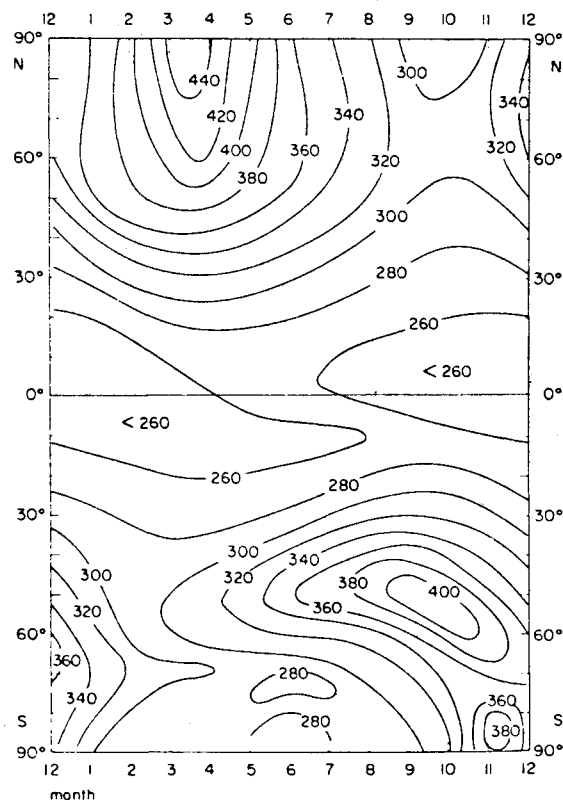


Fig. 1 Total ozone as function of season and latitude in both hemispheres [From Dütsch, (1971), based upon data by London (1963), and Stickse (1970)].

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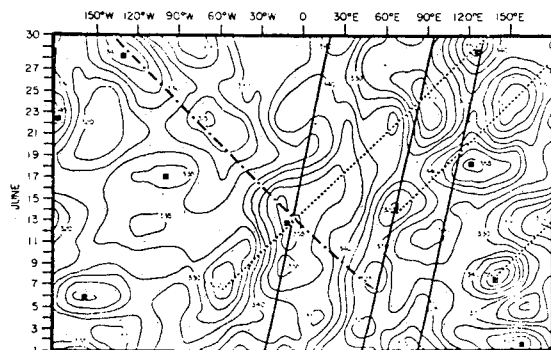


Fig. 2 Time-longitude variations of the total ozone (m-atm-cm) during June 1969 at 40°N. Squares denote prominent ozone maxima. Solid lines are phase speeds of waves progressing eastward, dotted and dashed-dotted lines denote possible group-velocity propagation [From Reiter and Lovill, (1973)].

(Fig. 3) (see Reiter 1971, Vol. 2, p. 198-211 for references). Major incursions of Mt. Agung debris into south polar regions, presumably also traveling in the major aerosol layer, occurred—nota bene—during December 1963 when, according to Fig. 1, meridional transport processes in the stratosphere are best developed (Figs. 4 and 5). It is questionable, however, whether the barrier to meridional transport processes that seems to exist near 60°S (Fig. 1) allows a regular and persistent formation of such an aerosol layer over the Antarctic continent.

#### Hemispheric Differences

Figure 6 presents one strong but “lopsided” ozone maximum over Antarctica in the Australian sector, whereas in the northern hemisphere the polar ozone maximum is better centered and a number of planetary waves, strengthened by the superposition of cyclone waves, are visible (Fig. 7). (Lovill (1972) showed that strong meridional gradients of total  $O_3$  exist near the position of the polar-front jet axis. The strength of this  $O_3$ -gradient is proportional to the wind speed in the jet stream, i.e. to the intensity of the jet circulation.) The  $O_3$  maximum over Antarctica in Fig. 6—showing winter conditions—is located over the Antarctic highlands. Reiter (1969, p. 155; 1971, p. 151) postulates a significant and persistent sinking motion throughout the stratosphere and lower troposphere over this region, forced by katabatic outflow of cold air along the slopes of the highland. Such flow conditions are not characteristic for the north polar regions.

The katabatic outflow also gives rise to a mid-winter maximum of  $O_3$ -concentrations in surface air of Antarctica (Fig. 8). In the northern hemisphere all present indications are for a spring maximum of surface  $O_3$ .

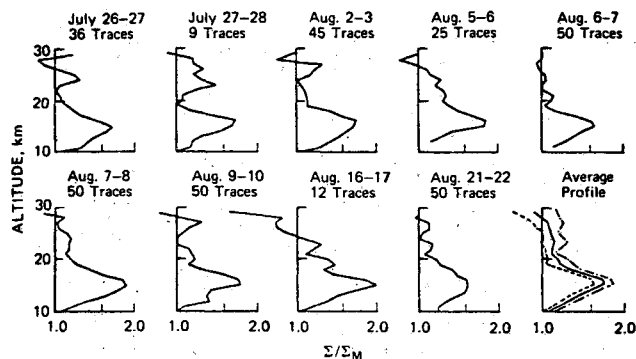


Fig. 3 Profiles of the scattering ratio  $\Sigma/\Sigma_M$  of lidar returns obtained during summer 1964 in Alaska [From Grams and Fiocco, (1967)].

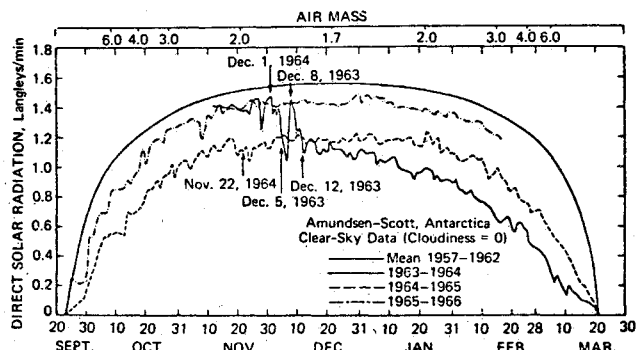


Fig. 4 Direct solar radiation data for Amundsen-Scott (South Pole) station [From Viebrock and Flowers, (1968)].

#### Secular Variations of Total $O_3$

There is some indication of a slow increase of total  $O_3$  in the 1960's (Fig. 9).  $NO_x$  created by nuclear blasts had no visible effect on such a trend. As yet it is not clear whether volcanic eruptions, nuclear blasts or solar activity are the primary causes for such long-term trends in  $O_3$ . It should be pointed out that Mastenbrook notes a secular increase of  $H_2O$  vapor in the stratosphere in the 1960's (Fig. 10). To my knowledge, the photochemical implications of this secular trend of  $H_2O$  on total  $O_3$  has not yet been explored. A possible interaction of these two trends should, however, be viewed as being of significant consequence to CIAP.

#### Short-term Fluctuations

In middle latitudes these variations are strongest in winter and spring and are caused by cyclone activity (Fig. 2). Vertical transport processes around jet streams of temperate latitudes produce the main “flushing” effect for the lower stratosphere (see Reiter, Vol. III, 1972). Since at these heights ozone is a quasiconservative atmospheric constituent, we have to presume that other atmospheric admixtures introduced into the atmosphere at these levels would be subject to the same transport and mixing processes.

One may attempt to estimate qualitatively the effectiveness of such mixing processes (for more quantitative

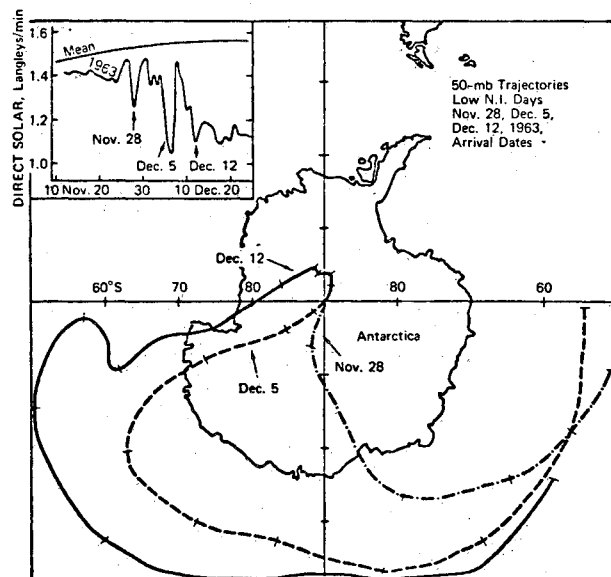


Fig. 5 Trajectories at 50 mb for air arriving at the South Pole on Nov. 28, Dec. 5, and Dec. 12, 1963. The diagram in the upper left shows the direct solar radiation at the South Pole on these dates [From Viebrock and Flowers, (1968)].

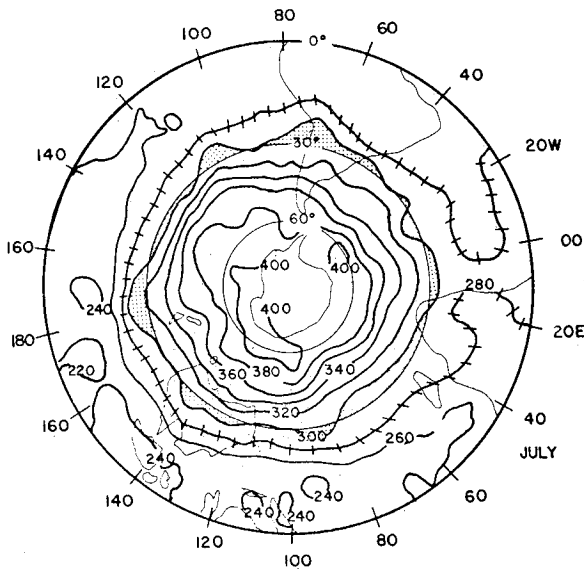


Fig. 6 Total ozone distribution in the Southern Hemisphere for July 1969. Total ozone values in m-atm-cm [From Lovill (1972)].

estimates one should turn to two- or three-dimensional stratospheric models). Figure 11 gives some clues. We note a general slope of the ozone peak from low to high latitudes, and from about 20 km to 16 km. (Equatorial peaks are located still higher. Photochemistry and upwelling motion in the equatorial stratosphere work together to create this effect.) The downward slope of the ozone peak coincides well with our notion on the behavior of the Junge aerosol layer. The meridional transport processes responsible for the ozone flux in this layer belong quite significantly to the standing planetary waves, so dominantly present in the northern hemisphere winter stratosphere, especially above the 20 km level. Below this level transport by transient cyclone waves gains importance. The "secondary maxima" on the spring ozone profiles of midlatitudes (Fig. 11) attest to the effectiveness of this transport by cyclone waves: ozone-rich air is transported from the stratosphere into the troposphere within the stable layer underneath the jet-stream (the so-called "jet-stream front"). Ozone-poor tropospheric air flows back into the stratosphere in the jet-core itself. It appears as a

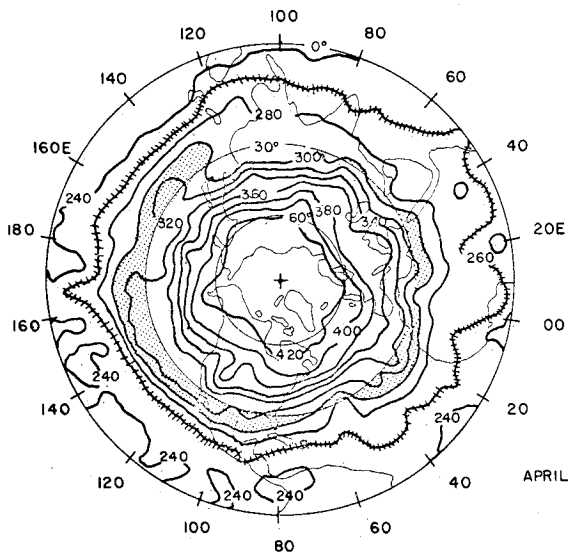


Fig. 7 Total ozone distribution in the Northern Hemisphere for April 1969. (Total ozone values in m-atm-cm) [From Lovill (1972)].

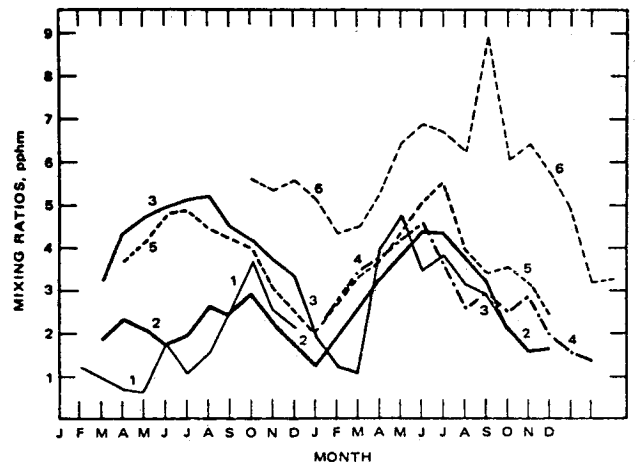


Fig. 8 Monthly mean surface ozone mixing ratios (in parts per hundred million) for (1) Dumont d'Urville, 1958; (2) Halley Bay, 1958-1959; (3) Little America, 1957-1958; (4) Hallett station, 1962; (5) Base King Baudouin, 1965-1966; (6) Amundsen-Scott Base, 1961-1962. [From Wisse and Meerburg, (1969)].

secondary ozone minimum on the middle- and high-latitude ozone profiles of spring in Fig. 11a near the 15-km level. This level (15 to 16 km) obviously marks the limit in altitude to which the rapid flushing of the lower stratosphere in middle latitudes near jet-stream systems proceeds. Stratospheric half-residence times (time required to remove  $\frac{1}{2}$  of the observed concentration) of atmospheric mass and pollutants in this region below 15 km in the vicinity of jet streams has to be assumed to be of the order of days to weeks, especially during the season of active jet stream systems, i.e. winter and spring. From evidence presented in Figure 1 we have to assume that this conclusion is valid for both hemispheres.

Above 15 km, and in the aerosol layer itself (15-20 km level, sloping upward from the pole towards the equator) half-residence times increase from weeks to months, per-

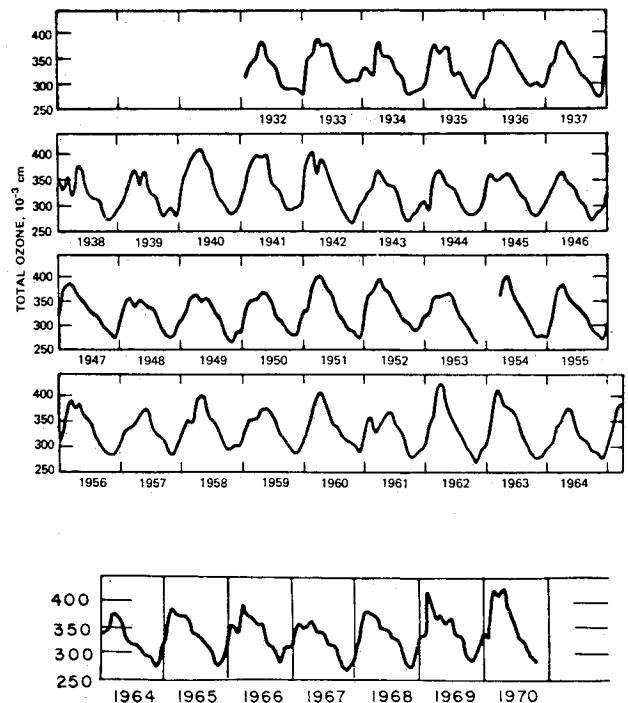


Fig. 9 Monthly mean total ozone amounts at Arosa, Switzerland (46.5°N, 9.4°E) [Upper Portion of diagram from Wallace and Newell (1966); lower portion: data courtesy of Dr. J. London].



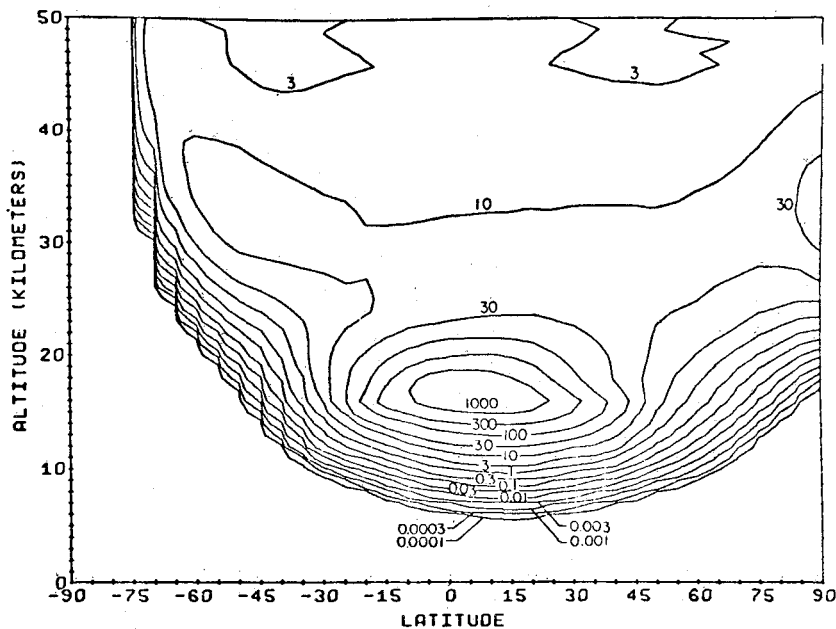


Fig. 13 Ratio of ozone production rate to ozone destruction rate according to Chapman mechanism [From Johnston (1973)].

#### Photochemistry of $O_3$ and the $NO_x$ Problem

Reiter (1971, p. 59) lists 36 chemical reactions in the upper atmosphere. All these reactions, in some way or other, have a bearing on the photochemistry of ozone. They include water vapor and other hydrogen compounds, but not the nitrogen compounds. Consideration of the latter makes the chemistry and photochemistry of the upper atmosphere considerably more complicated. Not only is nitrogen important in the three-body collision postulated in the Chapman equations of ozone formation, but the oxides of nitrogen play a very important role in the catalytic destruction of ozone. The chemistry and photochemistry of  $NO_x$ , therefore, will have to be considered very carefully when assessing the possible impact of the SST on the environment.

In Fig. 13 the ratio of  $O_3$  formation to  $O_3$  destruction is shown for the Chapman equations. It is quite obvious that these equations alone cannot account for a photochemical equilibrium, since in the lower stratosphere approximately 1000-times more ozone would be produced than destroyed. Introduction of  $4.2 \times 10^9$  molecules/cm<sup>3</sup> of  $NO_x$  through-

out the stratosphere, according to Fig. 14, would bring about a rather close photochemical equilibrium of ozone.  $NO_x$ , therefore, may appear as a rather intriguing "solution" to the ozone problem. It has to be pointed out, however, that Fig. 14 has been derived for a motionless atmosphere. This does not conform to reality.

We know that  $O_3$  is moved through the lower stratosphere by atmospheric motion systems of a variety of time and distance scales. From studies of detailed  $O_3$ -measurements by balloon soundings, and from (isentropic) trajectory analyses, we find our conclusions confirmed that  $O_3$  in the lower stratosphere behaves almost as a conservative tracer. It, therefore, does *not* seem to undergo *appreciable* chemical or photochemical reactions at altitudes of 20 km or lower. Atmospheric motions, thus, appear to be in direct competition with  $NO_x$  in explaining the ozone distribution in the stratosphere.

Furthermore, it should be pointed out, that the *concentration* of  $4.2 \times 10^9$  molecules/cm<sup>3</sup> assumed by Johnston (1973) to be *uniform* throughout the stratosphere does *not* conform to *constant mixing ratio* in meteorological terms. Because pressure is decreasing with height, a *constant*

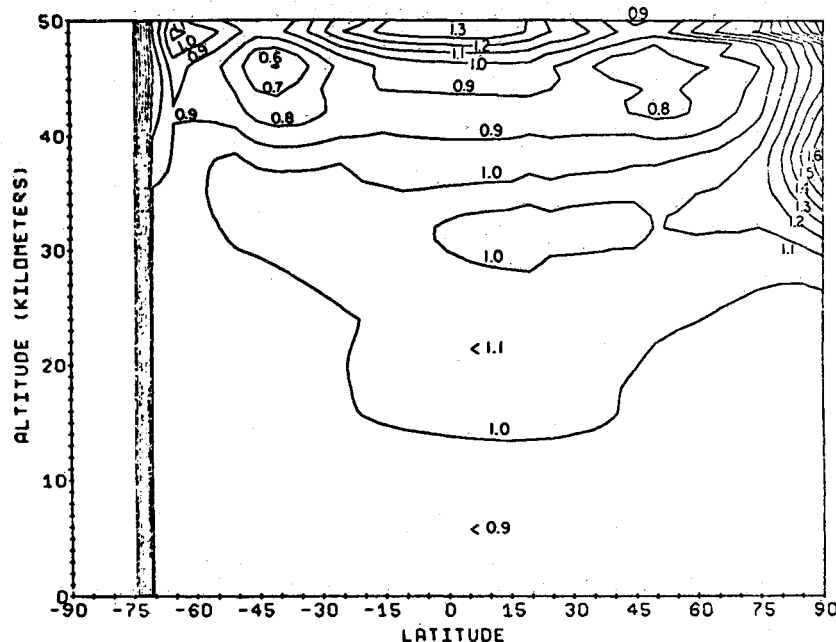


Fig. 14 Same as Fig. 13, except  $4.2 \times 10^9$  molecules/cm<sup>3</sup> of  $NO_x$  added. [From Johnston (1973)].

chemical concentration would mean mixing ratios (mg of  $\text{NO}_x$  per g of air) increasing linearly with decreasing pressure, or increasing with height as pressure decreases. Mixing ratio is conserved during atmospheric transport processes that occur *without* mixing of different air masses, but chemical concentration is not. This does not belittle the fact, that the chemistry and photochemistry of  $\text{NO}_x$  is of great importance in the ozone region. But so are atmospheric transport processes. Something must happen to the  $\text{NO}_x$  which is present in the atmosphere even nowadays before the onset of SST traffic, that makes it less effective in destroying  $\text{O}_3$  than would appear from Johnston's model. What are the natural sinks of  $\text{NO}_x$ ? Conversion to  $\text{HNO}_3$  seems plausible.  $\text{CH}_4$  may play a role in this conversion, so may the catalytic action of aerosols and ions of trace substances, about which we know hardly anything at all. There are, however, a lot of unsolved questions that make the removal rate of  $\text{NO}_x$  from the atmosphere an unknown quantity. Such removal possibilities will have to be studied in detail, however, if a credible estimate of the disposal of additional  $\text{NO}_x$  produced by "unclean" types of SST engines should be made.

### Radiation and $\text{O}_3$

Since  $\text{O}_3$  is generated photochemically in the stratosphere it is not only affected by, but also has a profound effect upon, the radiation fluxes that permeate the atmosphere. Photochemical models are based upon a certain balance between UV radiation fluxes and  $\text{O}_3$  concentrations (as well as the concentrations of other chemical species). The important questions arise: a) whether or not any changes in ozone concentrations in a particular layer of the stratosphere will alter the photochemistry of lower layers in a significant way; b) whether or not any changes in total stratospheric ozone will change the UV spectrum and its intensity received at the earth's surface. The biological significance of such changes will have to be assessed separately; and, c) whether or not changes in atmospheric  $\text{O}_3$  concentrations may alter the radiation fluxes through the atmosphere, hence the heating rates, and thereby cause changes in climatic regimes.

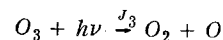
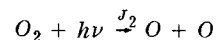
A serious problem in trying to answer these questions arises from the fact that the transmissivity of the atmosphere as a function of radiation wave length is not precisely known, nor is the original solar brightness (as a function of wavelength) outside the earth's atmosphere. The problem is compounded by the fact that the sun is a variable star, with significant changes occurring in its UV spectrum between sun spot maximum and minimum. The composition of the atmosphere, especially with respect to aerosols and other trace materials, also cannot be considered as constant with time and space.

In the wave lengths effected strongly by ozone, centered at 210 nm, values of energy fluxes computed by Brewer and Wilson may be too low by a factor of four. Whether solar radiation changes cause changes in total ozone has not yet been determined unambiguously. Radiation changes of the sun are quite significant, especially in the X-ray region of the spectrum. The 11-year sunspot cycle and the 27-day rotation cycle play major roles in the solar radiation variability. One will have to allow for these changes in solar intensities when one attempts to answer questions a, b, and c.

In estimating the balance between radiation intensities at a given level, and ozone generation and destruction, one runs again into the problem of the inadequacies of the simple Chapman model, and the relative effects of atmospheric motions and more complex photochemical reactions.

Between 296 nm and 222 nm there is almost complete extinction of UV radiation because of the Hartley continuum of ozone. This spectral feature, the so-called "Hart-

ley Darkness," has important consequences on the dissociation rates  $J_2$  and  $J_3$  which govern the photochemical reactions



Suppose the amount of  $\text{O}_3$  were reduced above level  $z$ . The Hartley Darkness, in this case, will brighten at level  $z$ , and due to the greater UV intensity the rate  $J_2$  will increase at level  $z$ .  $J_3$ , on the other hand, is dominated mainly by the optically thin Chappius band and will remain essentially unchanged. Thus  $J_2/J_3$  will increase and so will the local  $\text{O}_3$  concentration at level  $z$ . Thus, a decrease or increase of ozone above level  $z$  will result in an increase or decrease, respectively, of ozone at level  $z$ . The total ozone, integrated over an air column from the ground to the top of the atmosphere, therefore, will remain essentially unchanged.

This conclusion helps to answer questions a and b: a) A loss of  $\text{O}_3$  above a certain level is expected to yield an increase at and below this level, and b) The net effect of these changes in the ozone layer—provided that they do not affect the whole ozone region, i.e. the whole stratosphere, in the same sense—will be rather negligible at the earth's surface.

This compensating effect, brought about by the Hartley continuum of ozone, makes it difficult to assess the virtues of one photochemical model of the atmosphere versus those of another. It should also be pointed out that there are still uncertainties in the dissociation rates  $J_2$  and  $J_3$ .

The heating rates in the stratosphere by  $\text{O}_3$  in the UV and visible parts of the spectrum depend among other things on the mixing ratio of  $\text{O}_3$  and on the photo-dissociation rate,  $J_3$ . If, for some reason, the  $\text{O}_3$  concentration were decreased above a certain level  $z$ , the heating rate  $T$  should decrease because of the decreased ozone mixing ratio. Below level  $z$  the ozone mixing ratio would increase, so would  $T$ , because of the mixing-ratio increase, but also because of the increased radiation energy that enters into the dissociation rate  $J_3$ .

Thus, it appears that reduction of ozone in a confined layer of the stratosphere could lead to a slow destabilization because of the UV and visible-light heating rates. It will yet have to be estimated, probably from numerical models, whether this destabilization will proceed rapidly enough to enhance turbulent exchange in a significant way. This, again, would have to be considered a "self-healing" characteristic of the ozone layer.

The infrared radiation fluxes also tend to offset slightly the effects of UV and visible light on the heating rates. A decrease in  $\text{O}_3$  concentration would lead to a decrease of infrared cooling in the stratosphere.

The presence of aerosols may change the radiation fluxes in the ozone layer significantly. Not enough is known about the optical properties and the distribution of stratospheric aerosols to permit detailed treatment of their effects on heating rates. In the infrared region a better knowledge of the refractive index of aerosols is even more crucial than in the UV and visible portion of the solar radiation spectrum.

Obviously, not all ongoing studies in the fields of stratospheric chemistry, photochemistry and radiation physics can be mentioned here. For more detail, and for an exhaustive summary of literature references upon which the statements made above are based, the reader is referred to CIAP Monograph Vol. 1, "The Natural Stratosphere" which is presently in preparation and will be available for general distribution early in 1974.

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J. AIRCRAFT

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# Engine Exhaust Emission Levels

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As a part of the U.S. Department of Transportation's Climatic Impact Assessment Program (CIAP), the exhaust emission products from a YJ93-GE-3 afterburning turbojet engine were measured under simulated flight conditions. The results show that the emission indices for both the formation of carbon monoxide (CO) and total unburned hydrocarbons (THC) increase with increasing altitude at constant Mach number; both decrease with increasing Mach number at constant altitude. The results also show that the emission index for nitric oxide (NO) formation increases with increasing Mach number at constant altitude and decreases with increasing altitude at constant Mach number.

## Introduction

THE United States Department of Transportation is conducting a Climatic Impact Assessment Program (CIAP). The objective of this program, as defined by its director, Dr. A. J. Grobeger, is "to assess, by report in 1974, the impact of the environmental and meteorological changes due to a world high-altitude vehicle fleet as projected to 1990."<sup>1</sup> During the initial planning for CIAP the primary concern was directed toward the impact of supersonic aircraft. However, as reported by W. B. Beckwith<sup>2</sup> and J. F. Leach et al.<sup>3</sup>, it has become apparent that there is already a significant number of subsonic aircraft flights in the stratosphere. Consequently, CIAP is now concerned with any and all vehicles that may be operating in the stratosphere in 1990.

The measurement of the exhaust emissions from several existing engines under simulated flight conditions is included in the various projects of CIAP. This paper presents and discusses some of the results of one of the measurement tests.

## Test Description

The test covered herein was conducted in July 1972 in the Engine Test Facility at the U.S. Air Force Arnold Engineering Development Center (AEDC) near Tullahoma, Tennessee. A General Electric YJ93-GE-3 engine, originally used in the XB-70 supersonic bomber, was used for the test. The YJ93-GE-3 engine is an afterburning turbojet engine designed for cruise at a Mach 3 flight speed.

Exhaust emission measurements were made over a range of altitudes from sea level to 75,000 ft and a range of flight speeds from zero to Mach 2.6. Two power settings were used at almost all altitudes. These power settings were: 1) maximum nonafterburning or military power, and 2) partial afterburning power simulating a supersonic cruise condition. At sea level a variable engine rpm test was conducted. At several conditions variable afterburner fuel/air ratio tests were run.

The test was conducted at AEDC in altitude cell J-2, which is 20 ft in diam. In the cell, the engine inlet and exhaust conditions are controlled to simulate the conditions of temperature, pressure, and airflow equivalent to what the engine would experience in flight.

On-line instrumentation was used to measure the exhaust products of interest in this study. The exhaust gas sampling system was designed insofar as practicable to meet the requirements of SAE Aerospace Recommended Practice (ARP) 1256, "Procedures for the Continuous Sampling and Measurement of Gaseous Emissions from Aircraft Turbine Engines." A single-point sampling probe

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Index categories: Airbreathing Engine Testing; Airbreathing Propulsion, Subsonic and Supersonic.

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