Stratospheric Effects from Aviation

Anthony J. Broderick
Federal Aviation Administration, Washington, D.C.

Introduction

UNTIL quite recently, interest in atmospheric ozone was largely academic. More or less regular measurements of this minor constituent of the air, about 90% of which exists in the stratosphere, have been made since the mid-1920's. But, during the widely publicized public debate that culminated in the withdrawal of federal financial support for the development of a commercial supersonic transport (SST), McDonald[1] pointed out that a small reduction in average atmospheric ozone concentration would be expected to lead to an increase in the incidence of skin cancer. At that time, it had already been claimed[2] that a fleet of 500 U.S. SST's would likely reduce the average ozone amount by a few percent. Fears were also expressed[3] about that time that persistent stratospheric contrails would result from SST operations, effecting a potentially significant cooling of the Earth's surface.

In 1971, the U.S. Congress decided to terminate commercial SST development funding. But the continuing Anglo-French Concorde and Soviet TU-144 development programs, coupled with the general environmental concerns of the period, gave impetus to a cooperative international scientific investigation of the potential seriousness of the environmental problems that had become associated with supersonic flight. Quite early in the formative stages of this effort, it was acknowledged by most scientists that SST's would form contrails in the stratosphere only rarely, because of that region's extremely low (typically only a few percent) relative humidity. It was also pointed out that the earlier calculations concerning ozone reduction by water vapor had been based on unverified assumptions about chemical reaction rates. But the "SST-ozone" problem had not been discounted, since independent work of Crutzen[4] and Johnston[5] had laid the foundation for a new source of environmental concern: it was theorized that oxides of nitrogen, present in small amounts in all aircraft exhaust, could reduce ozone. Since material injected directly into the stratosphere resides there for months to years (this "residence time" increasing with height), it was argued that the stratospheric buildup of oxides of nitrogen (NO_x) from SST's would result in a considerably greater ozone reduction than had earlier[2] been attributed to water vapor emissions. It was on investigation of this phenomenon that the subsequent international efforts were centered.

In the United States, the Department of Transportation Climatic Impact Assessment Program (CIAP) was initiated in August 1971, and a parallel independent effort by the National Academy of Sciences (NAS) was organized shortly thereafter. Technical work on these comprehensive programs was completed by late 1974, and the final reports of the CIAP[6] and NAS[7] studies were released in early 1975. In the United Kingdom, the Committee on Meteorological Effects of Stratospheric Aircraft (COMESA) was formed and conducted a similar study from 1972 to 1975, its report[8] being released some months after the U.S. reports. During this time, the Comite sur les Consequences des Vols Stratospheriques (COVOS) in France also conducted a major study, its final report[9] having recently been made available. In general, the results of these studies were in good agreement. Table 1 shows a summary of their results concerning ozone reduction from SST operations. Note that, to facilitate comparison, we have normalized emission factors (18 g of NO_x as NO_2 per kg of fuel burned), cruise fuel consumption (19,100 kg per hour), flight altitude (16.5 km), and annual cruise time (4.4 hours per day in the stratosphere, 365 days per year—an implicit fleet average aircraft utilization on the order of 8 to 9 hours per day). These values were those concluded by the CIAP study to be realistic, and we characterize the aircraft in this discussion as "Concorde-like" to distinguish it from radically different designs (such as that of the 1971 Boeing 2707). We also intend this "normalized" aircraft to broadly represent the TU-144, insofar as its reported characteristics[10] are quite similar.

These studies concerning "SST-ozone" effects carried major implications for subsonic aircraft operations in their final reports. This was particularly true for the CIAP and NAS studies; the COMESA report was not as comprehensive in this regard, and the COVOS report presented no quantitative estimates of the subsonic aircraft effect on ozone. Note that the delineation between subsonic and supersonic flight is actually artificial—it is flight altitude that is of concern; much long-range subsonic flight does and will increasingly occur in the lower stratosphere. Extrapolations of high-altitude (20 km) SST effects to as low as 9 km indicated that even present subsonic aircraft flights would reduce ozone levels. The unit reduction of ozone on a "per-aircraft" basis was calculated to be considerably smaller than that for SST's, however, primarily because the "residence time" of

Anthony J. Broderick received his B.S. in Physics from St. Bonaventure University in 1964. From 1964–1970 he worked in a program management capacity for several electro-optical instrumentation companies. In 1970, he joined the U.S. Department of Transportation, specializing in air quality studies. His work on the Department of Transportation Climatic Impact Assessment Program (1970–1974) and the Federal Aviation Administration's High Altitude Pollution Program (1975–present) has occupied the bulk of his time ever since. He sits on a number of Federal Committees, including the Interagency Committee for Stratospheric Ozone Protection of the Environmental Protection Agency, the Working Group on Stratospheric Monitoring of the National Oceanic and Atmospheric Administration, and the Committee on International Environmental Affairs of the Department of State. Mr. Broderick has been an Adviser to U.S. Delegations to the United Nations Environment Programme and the U.S.-U.S.S.R. Bilateral Agreement on Environmental Protection and is Alternate U.S. Member of the International Civil Aviation Organization's Committee on Aircraft Engine Emissions.

Presented as Paper 77-799 at the AIAA/SAE 13th Propulsion Conference, Orlando, Fla., July 11-13, 1977; submitted Aug. 31, 1977; revision received May 3, 1978.

Index category: Environmental Effects.

Table 1 Size of "Concorde-like" SST fleet required to reduce average Northern Hemispheric ozone concentrations by 0.5% after several years of continuous operation

Name of study	Number of aircraft
CIAP[6]	120
NAS[7]	80
COMESA[8]	440
COVOS[9]	330

Table 2 Estimated Northern Hemisphere ozone reductions from various postulated subsonic aircraft fleets for different studies

Study	Percent Northern Hemisphere ozone reduction
"1974 CIAP Fleet":	
CIAP[6]	0.09
NAS[7]	0.10
COMESA[8]	0.24
COVOS[9]	Not available
100 "747-SP-type" aircraft:	
CIAP[6]	0.08
NAS[7]	0.16
COMESA[8]	Not available
COVOS[9]	Not available

pollutants injected at such low altitudes was relatively short, and high-altitude stratospheric NO_x concentrations would not be perturbed so severely as in the case of SST flight.

The subsonic aircraft results are shown in Table 2 (normalized by us for clarity, as with Table 1). The characteristics assumed for the "1974" subsonic fleet (called the "present" subsonic fleet by CIAP) were as follows[6]: 1217 "B-707-like" aircraft, with cruise NO_x emissions of 6 g/kg of fuel burned, average of 5245 kg/h cruise fuel consumption; 232 "DC-10/L-1011-type" aircraft with cruise NO_x emissions of 15 g/kg of fuel burned, average of 7482 kg/h cruise fuel consumption; 232 "B-747-type" aircraft with cruise NO_x emissions of 15 g/kg of fuel burned, average of 10,219 kg/h of cruise fuel consumption; all cruising at 11 km for 5.4 hours per day, 365 days per year. For the 747-SP-type aircraft, B-747-type characteristics were assumed, but the cruise altitude assumed was 13.5 km. This produced a major difference in estimated ozone reduction, as shown in Table 2.

Practically speaking, the implications of the findings just summarized were actually far more serious in the near term for the subsonic aircraft fleet than for supersonic aircraft, since the possibility was thus raised that major combustion system redesigns would soon need to be accomplished for a large number of aircraft. At a CIAP-estimated cost of $50 million for research, development, and certification of such a system, plus added hardware and maintenance costs, the upper-bound estimate of the total investment required to keep stratospheric ozone reduction for the CIAP-estimated fleet below 0.5% by about 1990 was placed at about $7 billion (1976 dollars) by one respected airline executive.[11] This was nearly an order of magnitude more than the investment similarly calculated to be required to insure the same level of environmental protection from the CIAP-estimated "upper-bound" SST fleet of that time. Clearly, these matters deserved further study. The "upper-bound" fleet growth projection of CIAP for SST's was widely discounted as unrealistic, but the same was not so for the CIAP subsonic fleet projection (though it, too, is now believed to be overly optimistic).

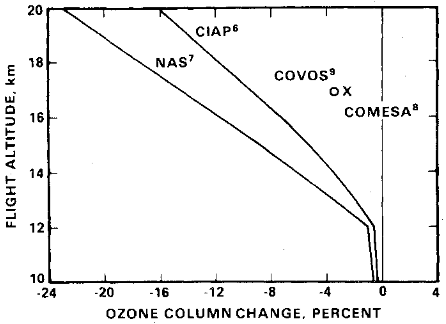

Fig. 1 Ozone column change results by various models. The injection rate assumed at each altitude is 1.23×10^9 kg NO_x (as NO_2) per year into a hemisphere. The NAS and CIAP curves are based on one-dimensional model results, adjusted to correspond to changes to be expected in the "hemisphere" (NAS) or "corridor" (CIAP). Points for COMESA (X) and COVOS (O) are calculated as explained in text (after Oliver et al.[15]).

Further U.S. studies were initiated in 1975[12] by the Federal Aviation Administration (FAA), under its High Altitude Pollution Program (HAPP). The HAPP objective is to reduce the scientific uncertainties in the foregoing predictions of aircraft effects, in order to develop a solid technical understanding of these factors as a basis for federal aviation policy development. Similar studies have also been continued in the United Kingdom and France, having been spurred by the Feb. 4, 1976, decision on Concorde[13] and the resulting Tripartite Agreement on Stratospheric Monitoring.[14] As a reference point, Fig. 1 summarizes graphically the results of the four "SST studies" briefly described in the foregoing.[15]

In this paper, we summarize the highlights of the current status of our understanding of the effects of high-altitude flight on ozone. Major factors in our perceptions of important atmospheric chemical reactions are reviewed, as are the relationships between aircraft and fluorocarbon-related stratospheric pollution. Gaps in our knowledge and some of the remaining scientific uncertainties are outlined and requirements for near-term research presented, along with some conclusions.

Considerations of Chemistry

In his classic work, Chapman[16] presented a theory explaining the presence of stratospheric ozone (O_3) which relied only on chemical reactions among O_3, oxygen atoms (O), and oxygen molecules (O_2) in the presence of a neutral third body, *M*, fueled by solar ultraviolet energy. This "pure oxygen" chemistry explained how ozone absorbed solar ultraviolet energy ($h\nu$), thereby shielding the Earth from most of this radiation:

$$O_2 + h\nu \rightarrow 2\,O \quad (h\nu \text{ below 240 nm}) \tag{1}$$

$$O + O_2 + M \rightarrow O_3 + M \tag{2}$$

$$O_3 + O \rightarrow O_2 + O_2 \tag{3}$$

$$O_3 + h\nu \rightarrow O_2 + O \quad (h\nu \text{ below 1140 nm}) \tag{4}$$

Ozone is destroyed by reaction (3) and reformed by reactions (1) and (2). Reaction (4) usually results in rapid reformation of ozone via reaction (2).

As measurements of ozone improved, it became clear that there was less ozone in the stratosphere than Chapman chemistry alone had explained, and "something else" must account for additional ozone destruction, as shown clearly by Johnston.[17] More than 20 years ago, Hampson, among others, proposed[18] that loss of ozone by reaction with active hydrogen ("water-based" or HO_x) species might be important in this regard, a theory later explored further by Hunt.[19] Logical extensions of this theory (flawed by erroneous assumptions, as mentioned earlier) led to the belief that stratospheric water from SST's would increase the natural HO_x fraction of ozone destruction, resulting in anthropogenic ozone decreases.[2] At about the same time that the early assumptions in water-based chemistry were reexamined, Crutzen[4] and Johnston[5] called attention to the NO_x problem, as mentioned earlier. Crudely, the NO_x and HO_x destruction of ozone may be summarized in simplified fashion as follows[7]:

$$O_3 + h\nu \rightarrow O_2 + O(^1D) \quad (h\nu \text{ below } 310\text{ nm}) \qquad (5)$$

$$O(^1D) + H_2O \rightarrow 2HO \qquad (6)$$

$$HO + O_3 \rightarrow HOO + O_2 \qquad (7)$$

$$HOO + O_3 \rightarrow HO + O_2 + O_2 \qquad (8)$$

$$HO + O \rightarrow H + O_2 \qquad (9)$$

$$HOO + O \rightarrow HO + O_2 \qquad (10)$$

$$H + O_3 \rightarrow HO + O_2 \qquad (11)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (12)$$

$$NO_2 + O \rightarrow NO + O_2 \qquad (13)$$

$$NO_2 + h\nu \rightarrow NO + O \qquad (14)$$

$$HO + NO_2 + M \rightarrow HNO_3 + M \qquad (15)$$

$$HNO_3 + h\nu \rightarrow HO + NO_2 \qquad (16)$$

Reactions (6) and (11) are the major sources of hydroxyl radicals (HO), which can also be formed by reaction of excited oxygen atoms $O(^1D)$ with methane (CH_4) and hydrogen molecules (H_2). In turn, the HO reacts with ozone [via reaction (7)] to produce perhydroxyl radicals (HOO). Both HOO and HO destroy ozone directly [e.g., reactions (7) and (8)] and indirectly by interference with reaction (2) [reactions (9) and (10)]. In the NO_x cycle, ozone destruction [i.e., in effect, reaction (3)] is catalyzed by reactions (12) and (13), with reaction (14) actually leading to small ozone production [via reaction (2)] in the upper troposphere and lower stratosphere. Reaction (15) breaks the chain of catalytic ozone destruction formed by reactions (12) and (13) and, since HNO_3 is quite water soluble, is the major "sink" mechanism for stratospheric oxides of nitrogen. (HNO_3 diffuses to the troposphere, where it can be removed by rain, usually after thousands of ozone molecules have been destroyed by the original NO_2 molecule.) Note that, although HNO_3 is in the stratosphere, reaction (16) serves to demonstrate that HNO_3 only "buffers" the NO_x destruction of ozone, since it may be photolyzed to yield NO_2 and HO.

In the course of these studies, it was recognized by Stolarski and Cicerone,[20] among others, that chlorine atoms could also "catalyze" ozone destruction by a mechanism parallel to the NO_x cycle:

$$Cl + O_3 \rightarrow ClO + O_2 \qquad (17)$$

$$ClO + O \rightarrow Cl + O_2 \qquad (18)$$

$$Cl + CH_4 \rightarrow HCl + CH_3 \qquad (19)$$

$$HO + HCl \rightarrow H_2O + Cl \qquad (20)$$

The initial importance of this ClO_x reaction cycle was thought to be with regard to the NASA Space Shuttle booster rocket, which contains HCl in its exhaust. Thus, chlorine would be released in an already "buffered" form (HCl) in the stratosphere, but reaction (20) would produce the catalysts for reactions (17) and (18) until the HCl was removed by rainout. Concern about this problem (50 Space Shuttle launches per year have been estimated[21] to reduce ozone by only a small fraction of 1%) was soon dwarfed by the now famous discovery of Molina and Rowland that vast amounts of stratospheric chlorine would be released by photolysis of fluorocarbons ($CFCl_3$ and CF_2Cl_2, sometimes referred to as "Freons" or F-11 and F-12, respectively). It is this latter source of ozone reduction that has dominated public attention and research for the past two years.

Clearly, a number of substances, natural and anthropogenic, are involved in ozone chemistry. When all of the known important reactions affecting stratospheric ozone are noted, they number around 100 or so and involve several dozen different chemical species, depending upon the details of the question one wishes to pursue. Two important general considerations deserve special attention: the complex competing relationships among the various ozone production and destruction reactions, and the uncertainties in our ability to quantitatively predict (i.e., mathematically "model") the system's behavior.

It is implicit in the foregoing that the natural HO_x, ClO_x, and NO_x systems must be "balanced" in the absence of anthropogenic influences, since there is only a fixed amount of ozone which represents the excess of ozone production over ozone loss by the Chapman system [reactions (1-4)]. In addition to being balanced, these systems are closely coupled, both directly and indirectly. For example, reaction (15) removes both HO and NO_2 from their respective catalytic ozone destruction cycles—reactions (7) and (8) and reactions (12) and (13). In the case of NO_2, for example, this slows down the speed of catalytic destruction of ozone, since the rate at which reaction (13) proceeds determines the amount of ozone destruction which will be realized. Reaction (13) is thus sometimes termed the "rate-limiting" step. But such a simplified view of things must be formed and interpreted only with great caution. In the simple catalytic cycles that we just described, such an analysis may be valid. But we are now beginning to recognize the complexity of the real atmospheric system. Not all of the important chemical subsystems are simple two-reaction mechanisms that proceed perhaps thousands of times before one of the constituents is removed (e.g., by $HO + NO_2 + M \rightarrow HNO_3$). Many of the reaction sequences involve half a dozen or more steps, with alternative reaction pathways at more than one point along the way. These more complex reaction sequences can be of major importance to the overall stratospheric ozone balance and do not lend themselves to simple analysis. Complex models must be brought to bear on the problem and their results carefully interpreted.

To resort, for the sake of example, to simple recitations of chemical coupling, consider that the following reactions are all of some importance in attempting to model stratospheric chemistry:

$$HO + NO_2 + M \rightarrow HNO_3 + M \qquad (15)$$

$$HO + HCl \rightarrow H_2O + Cl \qquad (20)$$

$$ClO + NO_2 + M \rightarrow ClONO_2 + M \qquad (21)$$

$$HOO + NO \rightarrow HO + NO_2 \qquad (22)$$

$$HO + HOO \rightarrow H_2O + O_2 \qquad (23)$$

$$NO + ClO \rightarrow NO_2 + Cl \qquad (24)$$

It is clear that, in addition to ClO_x, HO_x and NO_x directly reacting with ozone, the reaction of many forms of these species directly with each other may be of importance. The importance of reactions (15) and (20) was discussed earlier. Including reaction (21) is of major importance to those attempting to understand the effects of fluorocarbon release on ozone and is presently responsible for a reduction of the calculated ozone-destruction effectiveness of chlorofluorocarbons by about a factor of 2 (i.e., to 50% of what would be computed without this one reaction). Reaction (22) is important now because it had previously been thought to proceed at a rate approximately 20 times slower than was measured recently by Howard and Evenson.[22] Using the newly measured rate, the calculated effects of aircraft are reduced dramatically. (The rapid rate of this reaction also causes a re-evaluation of other important reaction rates that had earlier been estimated by interpreting experiments providing only data on ratios of the rate constants. Thus, the single measurement of Howard and Evenson has had a major effect on recommended values for several rate constants.) Reaction (23) terminates the HO_x catalytic ozone-destruction chain, but, since a balance among ozone-destruction mechanisms must be maintained, the recently accepted[23] slower rate for this reaction shifts a significant amount of calculated ozone reduction from NO_x to HO_x. Reaction (24) short-circuits both the NO_x cycle and the ClO_x cycle and is thus of importance to both.

With the kinds of complexity just described, simple uncertainty analyses should not be expected to produce meaningful answers. Indeed, it is instructive to note that the large uncertainties now recognized in our understanding of stratospheric chemistry were not explicitly recognized in any of the CIAP, NAS, COMESA, or COVOS reports discussed earlier. At the writing of both the CIAP and NAS studies, attention was focused on atmospheric transport uncertainty. In large part, this was due to a conceptual difficulty: one can measure the uncertainty of something relative to a value known to be "true," but if one does not know "the truth," how can one estimate the uncertainty of one's limited knowledge? To our knowledge, only one serious attempt at an uncertainty estimate has been published in the referenced literature, that by Duewer et al.[24] (For a complete discussion, one is also referred to Johnston and Nelson's commentary[25] on that paper and the accompanying reply by Duewer et al.[26])

Duewer et al. present a series of model experiments in an attempt to examine the sensitivity of model-calculated ozone reduction for a given stratospheric NO_x injection. The model did not include chlorine reactions and used the reaction-rate estimates and uncertainties assigned by Hampson and Garvin,[27] which were the basis of CIAP and NAS computations as well as the principal source of data for modeling studies under COMESA and COVOS. In brief, they found a startling sensitivity to uncertainties, even when only a small number of rates were considered. Carefully stressing that they did not really believe that it was realistic, they showed that, if one were to minimize the ozone-destroying effect of only three reactions (but remain within the bounds of uncertainty earlier established for each reaction by Hampson and Garvin), flight of "Concorde-like" (our term) SST aircraft would actually *in*crease ozone. Adopting the 1976 NAS-assumed[23] value for only one of these reactions [here denoted reaction (23)] caused a reduction in the calculated effectiveness of stratospheric NO_x by a factor of 2. Their conclusions pointed up the caution that must be used in this new area of atmospheric science and indicated that more accurate determinations of much of the existing chemical reaction-rate data are sorely needed for $HO_2 + HO_2$ and our reactions (8, 15, 22, and 23). The reaction $HOO + NO_2 \rightarrow HOONO_2$ was not included in this study and may also be of importance.

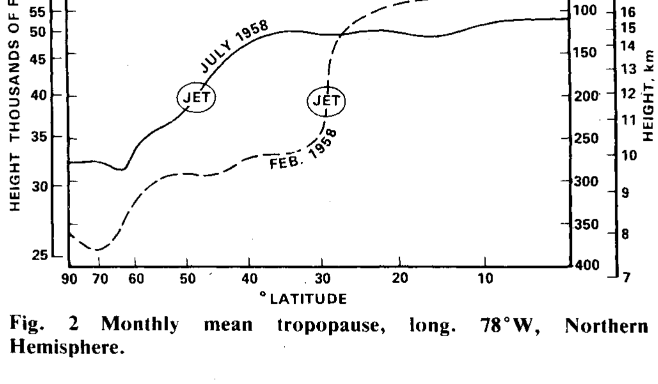

Fig. 2 Monthly mean tropopause, long. 78°W, Northern Hemisphere.

Transport Parameterizations

The study of aircraft effects on ozone is considerably different from that of fluorocarbons on ozone in regard to the importance of assumptions made about atmospheric transport. Briefly, fluorocarbons are inert at altitudes below about 30 km, above which they are photolyzed by solar ultraviolet radiation to produce chlorine. The length of time that it takes the average fluorocarbon molecule to reach this altitude is quite long, on the order of several decades at equilibrium. The active life of the chlorine atom in terms of ozone destruction, however, is much shorter, on the order of a couple of years. The former (upward) transport time results from the fact that surface releases must first mix uniformly in the lower atmosphere, slowly filling it up to some equilibrium level, before the material can reach upper stratospheric regions in significant amounts. Thus, an equilibrium amount of inert fluorocarbons is first realized in the lower atmosphere before it is achieved in the upper regions of the stratosphere. Once a chlorine atom is released by photolysis in this upper stratospheric chlorine-source region, however, an inverse situation exists. The atmosphere then fills from the top down. Since the chlorine is only active in the stratosphere (which constitutes about 10% of the atmospheric mass), it is removed relatively rapidly (on the order of 2-3 yr) to the troposphere and rained out (as HCl). The details of the upward and downward transport are of little importance. Whether it takes 90 or 100 years for the average fluorocarbon molecule to reach heights where it may be photolyzed is clearly irrelevant. Similarly, estimates of downward transport of such high-altitude injections (via photolysis) of chlorine have been made with an accuracy sufficient to establish the magnitude of chlorine destruction which will occur. For fluorocarbons, then, uncertainties in chemistry (and particularly in the validity of the assumption of their lower-atmospheric inertness) far outweigh any transport uncertainties, because of the altitude of the "injection" of the active species.

Such is not the case for aircraft-NO_x injections that take place between about 10 and 18 km. This is the region of the atmosphere where transport uncertainties are greatest (for consideration of average global-scale mixing). Here the tropopause is defined as the dividing line between stratosphere above and troposphere below, and its location is usually assigned to a well-defined inflection point in the lapse rate. Figure 2 shows a good example of why one cannot term the tropopause well behaved—it is highly variable in space and time.

Any model that seeks to assess the amount of ozone destruction associated with aircraft-NO_x injections must account for the rate at which the NO_x is removed (upward or downward) from the region of injection. Simplistically, on a global-average scale, upward transport from aircraft flight altitudes toward the region of high ozone concentration will result in ozone destruction, whereas downward or quasihorizontal transport has the opposite effect. Thus, transport uncertainties can place a fundamental limit on our quantitative understanding of aircraft effects.

Small-scale turbulence is one form of atmospheric motion which immediately comes to mind, but a proper analysis must also take into account mean meridional (north-south) motions, transient disturbances propagated upward from the troposphere (as from hurricanes), gravity waves, and quasistationary waves. Unfortunately, these motions do not operate independently, and again we are forced to employ mathematical models as predictive tools. Atmospheric-circulation models that are based on fundamental physical principles and explicitly treat time-dependent motions in all three dimensions are called general-circulation models (GCM's). Some GCM's have been shown to represent the major atmospheric processes reasonably well in the region of our interest, as described by Smagorinsky.[28] (An excellent review of the general field is also provided by the National Academy of Sciences.[29]) GCM's are, unfortunately, massive and complex constructs that require at least several days of dedicated effort to properly design, construct, and test before they can be used. Calibration tests have demonstrated that some of these models are able to simulate the behavior of atmospheric trace substances well enough to give confidence in their usefulness as predictive tools, and at least one (described in Ref. 30) is currently being used at the Geophysical Fluid Dynamics Laboratory to predict dispersion and transport of high-altitude aircraft exhaust.[31] The complexity of these models, however, coupled with their present inability to include many chemical reactions because of computational size limitations, naturally leads to a desire for physically and statistically valid simplifications. One example of a simplified three-dimensional model is the quasigeostropic spectral model of Cunnold et al.[32] The low spatial resolution of such models, however, gives cause[30] for concern over their ability to adequately simulate stratospheric behavior, since a significant amount of tropospheric kinetic energy is found on relatively small scales (above planetary wave number 6) and may significantly affect stratospheric circulation.

Another possible method to reduce computational complexity is to "average out" presumably less important motions, as is done in "two-dimensional" (2-D) and "one-dimensional" (1-D) models. The 2-D models explicitly treat only north-south and vertical motions, whereas 1-D models explicitly treat only the global-average vertical motion components.

Unfortunately, there has yet to be found an incontrovertibly "correct" basis for the "parameterizations" that must be employed in simplified models of any dimension. The more simplified the model, the more highly parameterized it must be, and the more cautious one must be in interpretation and use of its results. Simplified models are largely based on physical intuition and phenomenology.

The most widely used models for studies of aircraft effects have been the 1-D models. These are highly parameterized transport models, but because of their ability to explicitly include virtually all important chemistry, they have proven highly useful. Most employ a "vertical eddy-diffusion" formulation, which describes global- (or hemispheric-) average net vertical transport of substances and is based on an engineering fit of some kind to a trace-constituent concentration profile (the "tracer"). (The eddy-diffusion coefficient, empirically determined by a fit to actual data, when multiplied by the local mean tracer gradient, is equal to the local tracer flux.) An excellent description of the derivation of one such eddy-diffusion coefficient (*not* specifically designed to treat aircraft effects) is given in Appendix B of Ref. 23. (The "stretching," "smoothing," "adjusting," "fitting," "shifting," and "assuming" described therein are instructive, especially for their implications.)

Unless an eddy-diffusion coefficient is specifically derived from mathematical inversion of a truly inert atmospheric tracer (and none presently employed falls into this category), it is not consistent to continue to employ the diffusion coefficient after changing any chemical reaction subroutines that directly or indirectly affect the calculated concentration of the "tracer." For example, diffusion coefficients based on inversion of the concentration vs altitude profile of CH_4, when the assumed rate of reaction for HO + HOO was 2×10^{-10} $mol^{-1}\ cm^3\ s^{-1}$, are no longer strictly valid when the assumed reaction rate is changed to 2×10^{-11}. (Reaction with HO is an important upper-stratospheric loss mechanism for CH_4.) This point of internal inconsistency of the present approaches to 1-D modeling is apparently of little concern to most researchers. Since the implied uncertainties that result have never been explored explicitly, we again point out the need for caution in interpretation of the results.

More to the point, however, is the fact that there does not presently exist a data set that can be used to derive an adequate parameterization of transport in the 10- to 20-km region. This arises from the fact that the data set must be highly accurate, and a globally representative set acquired over at least several years must be available. In addition to those requirements, all significant chemical sources and sinks of the constituent must be known, in order not to falsely ascribe, for example, concentration decreases arising from unknown chemistry to rapid transport out of the region under study.

When all is said and done, the 1-D models must be regarded as extremely powerful tools, if properly used. They have been used to establish the likely importance of individual chemical reactions, the apparent sensitivity of stratospheric chemistry to uncertainties in reaction rates, and the approximate natural concentration of atmospheric trace constituents. Un-

Table 3 Effect of various chemistry changes on computed global ozone reductions for NO_x injections at 17 and 20 km

Change made	Computed ozone reduction	
	17 km	20 km
Start (1974)[42]	4.8	~11
New Chang eddy-diffusion[23]	5.4	12
($N_2O + h\nu$) changed from Bates and Hays[43] to Johnston and Selwyn[44]	5.3	12
Add "smog" reactions: $OH + CO_2$, $O + CH_4$, $H_2O_2 + O$, $HO_2 + h\nu$, $OH + CH_4$	4.8	11
$OH + HO_2$ ($2 \times 10^{-10} \rightarrow 2 \times 10^{-11}$)	2.1	6.4
$OH + HNO_3$ ($1.3 \times 10^{-13} \rightarrow 8.9 \times 10^{-14}$)	2.0	6.0
$NO_3 + h\nu$ [branching, $NO + O_2 \rightarrow$ $\frac{2}{3}(NO_2 + O)$, $\frac{1}{3}(NO + O_2)$]	1.6	5.2
$O(^1D)$ reactions with N_2O, N_2, O_2, CH_4, H_2O changed from Hampson and Garvin recommendations[27] to Streit et al. recommendations[45]	1.5	4.9
$OH + NO_2 + M$ (Tsang[46] → Anastasi et al.[47])	1.5	4.8
$HO_2 + HO_2$ [$3 \times 10^{-11} \exp(-500/T) \rightarrow 1.7 \times 10^{-11} \exp(-500/T)$]	1.2	4.3
$H_2O + O$ [$8 \times 10^{-11} \exp(-500/T) \rightarrow 3 \times 10^{-11}$]	1.2	4.2

fortunately, in the case of aircraft, the state of our knowledge (and the increasingly *de*creasing calculated sensitivity of stratospheric ozone to NO_x injections) forces us to increase our emphasis on the importance of developing higher-dimensional models. Although it is not clear that 2-D models are more accurate than 1-D models in this context, the ability in principle to treat transport in the tropopause region more completely is the principal justification for their use.

We have to date relied principally on the work of two groups for 2-D modeling studies in HAPP. The latest pertinent results of these efforts are described by Widhopf[33] and by Hidalgo and Crutzen.[34] Again, the principal difficulty with regard to transport is the lack of fully satisfactory means for establishing the validity of the models' calculated transport of trace constituents. In the case of 2-D models mentioned previously, mean meridional motions are inputs, and three sets of eddy-diffusion coefficients (north-south, vertical, and north-south/vertical coupling) must be employed, all of which must be, of course, mutually consistent. There is no well accepted set of such data, and, as in the case of 1-D models, considerable intuition must be relied upon. It is interesting to note, at this point, that the need for measured data is approximately the same for the validation of all models: 1-D models need the same raw data set as 3-D models, contrary to intuition, if they are ever to be "validated" to the same degree of confidence. The only difference is the degree of averaging of the raw data.

As stated earlier, one of the least understood aspects of stratospheric pollution studies is the exchange of air across the tropopause, i.e., between the stratosphere and the troposphere. Such exchange, in the downward direction, constitutes an important removal mechanism for pollutants injected or formed in the stratosphere (e.g., radioactive debris or HNO_3) and, in the upward direction, can be a source of stratospheric pollutants (e.g., NO_x formation from N_2O). It is appropriate to discuss this matter more fully.

A direct calculation of stratospheric-tropospheric mass exchange is possible in principle if one has a knowledge of the distributions of vertical velocity and concentration. But it is almost impossible to directly observe vertical velocity over the large scales of transport which are important, and so its value has to be inferred by calculations using synoptic meteorological data. Unfortunately, the known techniques of doing so (e.g., kinematic method, adiabatic method, vorticity method) are all prone to large errors. If one can tag and follow air parcels in the course of their motion, greater accuracy could be achieved in determining stratospheric-tropospheric exchange.

Relatively inert tracers that remain in suspension in air can be expected to move with the parcels (unless mixing takes place) and will be subject to the same physical constraints as the parcels. Examples of such tracers are ozone mixing ratio (in the lower stratosphere) and radioactive "fallout." Any conservative physical property of the parcel can also be used as a "tracer" with equal effectiveness, in principle. One such property is the so-called potential vorticity, which is conserved during isentropic motion.

Mathematically, potential vorticity is a product of the measures of the resistance of an air parcel to vertical and horizontal displacement. Hence, heating above and cooling below (i.e., increased vertical stability) increase the potential vorticity. Thus the heating source of the ozone layer generates large values of potential vorticity in the stratosphere. Conversely, heating from below decreases potential vorticity. This is the case for the troposphere, which is heated by the Earth's surface.

Reed,[35] and Reed and Danielsen,[36] have shown that the stratospheric values of potential vorticity exceed the tropospheric values by one, two, or three orders of magnitude. This circumstance can be used to trace intrusions of stratospheric air into the troposphere and vice versa; i.e., since each air parcel conserves its value of potential vorticity, the air of stratospheric origin can be readily identified even after it is considered part of the troposphere.

Danielsen,[37] using isentropic analyses, studied stratospheric-tropospheric exchange. He showed, from in situ measurements of radioactive fallout, that the tropopause "folding" phenomenon was predictable, that it accompanied large-scale cyclogenesis, and that the stratospheric air could be identified by both radioactivity and potential vorticity. Later[38] he established that the radioactivity and ozone measurements were well correlated. Thus, potential vorticity, a purely meteorological quantity, can be used to analyze stratospheric-tropospheric exchange without incurring the large expense of making synoptic observations of ozone or radioactivity.

Present Understanding

Tables 1 and 2 and Fig. 1 summarized the status of understanding of aircraft effects on ozone derived from the CIAP, NAS, COMESA, and COVOS studies. Since the publication of those four reports, little additional information has been gained which permits major modification to the approximations that they employed to represent atmospheric transport. The most detailed work performed in this respect was mentioned earlier (see Appendix B of Ref. 23), and Chang et al.[39] have adopted this slightly modified vertical transport estimate in their modeling efforts. What is significant, however, are the major modifications to our understanding of aircraft effects on ozone which have arisen in the chemistry area. These changes involve two principal factors: a better appreciation of the uncertainties in our knowledge and a reduction in this uncertainty for several chemical reaction rates by virtue of their measurement in the laboratory; and the discovery of the importance of chlorine in the stratosphere, with its direct and indirect influences on calculations of aircraft effects.

To best understand the impact of changes that have taken place in chemistry, Table 3 lists 11 individual changes that have been made[40,41] to one model discussed by Chang and Johnston.[42] After each change, we note (to only two significant figures) the newly computed global reduction in ozone which is calculated for a NO_x injection of 2000 mol cm^{-3} s^{-1} (2.5×10^9 kg/yr) into a 1-km-thick layer centered at 17 or 20 km.

Note that the data in Table 3 still do not directly address stratospheric chlorine, which is now believed to be naturally present at a level of about one part per billion (1 ppb) in the stratosphere, and is increasing because of photolysis of fluorocarbons released at the surface in the past decade or so. Including about 1 ppb of chlorine in the preceding model reduces the ozone reduction for the respective 17- and 20-km NO_x injections from about 1.2% and 4.2% to about 0.7% and 3.3%, respectively. This arises from the slowing down of the NO_x catalytic cycle [reactions (12) and (13)] by reaction (24). In turn, reaction (21), when subsequently included, tends to increase somewhat the effects of NO_x catalytic cycle again, since ClO is tied up as $ClONO_2$, thereby slowing reaction (24) relative to reaction (12). [Although reaction (24) actually reduces the amount of NO_x available for ozone destruction, the increased speed of reaction (12) more than compensates for this.]

Increasing amounts of chlorine in the stratosphere are physically inevitable for the next decade or so, since the fluorocarbons that will cause them have already been released. (Even if stopped instantly, fluorocarbon releases of the past will act to increase the stratospheric chlorine content for about a decade as they are slowly transported upwards, as discussed in the earlier section on transport.) Realistically, it is likely that the chlorine content of the stratosphere will increase for a longer period, because of the economic and political difficulties in developing and implementing adequate substitutes on a world-wide scale. Thus, any model of NO_x

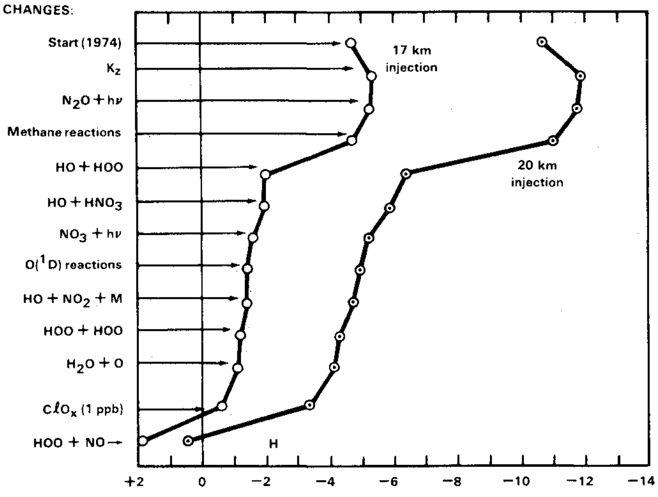

Fig. 3 Effects of changes in model assumptions (ordinate) on calculated global-ozone reduction (abscissa) for a NO_x injection of 2.46×10^{12} g/y at indicated altitude (after Chang[42]).

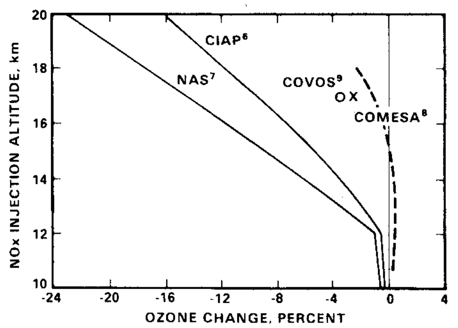

Fig. 4 Same as Fig. 1, but dashed line broadly represents results of later calculations before accounting for the presence of stratospheric chlorine and the recently measured[22] fast rate for HOO + NO (see text and Fig. 1; after Oliver et al.[15]).

effects from aircraft injections cannot be realistic without incorporating chlorine effects. But it is difficult to gage future stratospheric chlorine contents accurately, and about all that can intelligently be said is that, for about the next 30 years at the very least, the effects of stratospheric chlorine will reduce the sensitivity of stratospheric ozone to aircraft injections of NO_x.

Before discussing Fig. 3, it is important to recall the comments in the earlier section on chemistry about the major significance of Howard and Evenson's recent determination[22] of the very rapid rate for reaction (22) (HOO + NO) and its implications. The magnitude of this effect is clear from examination of Fig. 3, which presents a graphic display of the data in Table 3 and extends the data to include consideration of stratospheric chlorine and the new data of Howard and Evenson. Figure 3 is intended to illustrate how one model[42] has been affected by knowledge gained in the past two or three years. It is essential (and this cannot be stressed enough) that we remember that there are still major uncertainties and, likely, real gaps in our knowledge. It is possible, even likely, that some of the "revised" data shown on Table 3 and Fig. 3 will be revised once again. In addition, the transport coefficient used in the model can have a major impact on its output. If the "Hunten"[48] eddy-diffusion coefficient is used in this model, the 20-km injection of Fig. 3 is calculated to reduce ozone (as shown by the letter H on Fig. 3), and the production of ozone by 17-km NO_x injections is reduced by about a factor of 4. Nevertheless, one cannot escape the fact that, since the completion of the CIAP, NAS, COMESA, and COVOS studies, new data indicate strongly that the ozone layer is markedly less sensitive to injections of NO_x by SST's than was concluded by these studies. At this point, the sign of the effect (i.e., ozone *de*crease or *in*crease?) is not at all clear.

With regard to subsonic aircraft, much the same can be said, although perhaps with more confidence. Figure 4 is essentially the same as Fig. 1, but we have added a curve that is broadly representative of the results of studies recently described by Widhopf,[33] Hidalgo and Crutzen,[34] and those at the Lawrence Livermore Laboratory.[40] These results have recently been well summarized in detail by Oliver et al.[15], and do *not* account for chlorine in the stratosphere or for the changes required to take account of the results of Howard and Evenson.[22] It is clear, however, that, according to our best present understanding, the effect of subsonic (i.e., below about 15 km) flight is not such as to cause ozone reduction, but might even be to enhance ozone slightly. Considering the implications of including effects of chlorine and the results of Howard and Evenson on the conclusions implicit from Fig. 4, the case against ozone reduction resulting from subsonic aircraft operations is considerably strengthened.

Uncertainty Predictions

It was mentioned earlier that an analysis of the uncertainties in model predictions of effects on ozone is difficult. One approach that has proven quite useful is that of Duewer et al.[24] discussed briefly in the section on chemistry. An inherent difficulty in any such attempt is the extreme complexity of the models with which we must deal. They may involve about 100 nonlinearly coupled reactions and dozens of different reacting species. What we really seek is a method of assigning realistic probability distributions to the error inherent in each input parameter. But these are *not* solely random errors. They also include systematic errors, not readily subject to assignment of error bounds. Inductive reasoning simply does not lend itself to straightforward error analysis.

Whereas Duewer et al. used intuition and a painstaking trial-and-error process in their analysis, Stolarski et al.[49] have performed a different analysis of errors (in this case, for fluorocarbon studies, but the general results and methodology are applicable to NO_x studies). They provide results in reasonable agreement with the brief analysis of uncertainties described in Ref. 23. They employed a 1-D model and assigned each reaction rate an uncertainty based largely on estimates of Hampson and Garvin.[27] The form of this assigned uncertainty was that of a log-normal distribution. They then performed several hundred model runs, with each of the 48 reaction rates being individually varied according to its assigned probability distribution and a Monte Carlo scheme employed to propagate the uncertainties through the entire model calculation. They concluded that a reduction of all rate measurement uncertainties to ±15% would result in an overall uncertainty of ±30% for a given calculation of ozone reduction, but that, with present uncertainties, "the correspondence to a Gaussian is better on the high side, where a one-sigma uncertainty of a factor of 2 is found." Intuitively we would expect a somewhat greater uncertainty on the low side (i.e., less ozone reduction for a given ClO_x injection), since, after all, less than zero ozone reduction is possible, but Ref. 49 gives no further details.

The approach of Stolarski et al.[49] appears at first glance to be more rigorous than that of Duewer et al.[24], but we question the validity of assigning probability distributions, as they have done, to uncertainty estimates that are almost surely in error for systematic reasons and not for reasons of a lack of measurement precision. With the assumption that they have employed, what does the answer really mean from a practical viewpoint? We believe both methods to be useful, especially as tools for management of scientific programs that attempt to assess the stratospheric impact of various anthropogenic emissions.

Although the preceding uncertainty studies are useful as management tools, we believe that, given the present lack of measured data on atmospheric parameters that affect ozone (including ozone itself) and the inductive nature of the fundamental problem under investigation, neither study can be said to have given a true measure of uncertainty (nor do they so claim). This is dramatically borne out by the analysis of Duewer et al. which showed the impossibility of ruling out even highly improbable combinations of assumed reaction rates by comparing model predictions with available atmospheric measurements. The precision and accuracy of the existing field-measurement data, coupled with the inherent (and frustrating) natural variability of meteorological parameters, render such attempts useless except to point out grossly erroneous formulations. This capability to "bound" the problem should not be treated lightly, however, as it can occasionally prove quite useful. An example of an analysis of atmospheric measurements which proved useful in that manner is described by Ackerman et al.[50]

Gaps to be Filled

We are able to identify a number of significant areas of uncertainty which, among others, must be addressed if our understanding of the effects of aircraft NO_x emissions on stratospheric ozone is to be on a solid foundation. These are outlined below, in no particular order of importance.

There is an essential need for an improved atmospheric data base, for use in testing and calibrating atmospheric models. For aircraft studies, we particularly need data on the relationships among various individual species of the NO_x family in the presence of ozone. This calls for truly simultaneous (in space and time) measurements of the NO_x constituents and ozone at various latitudes and seasons, as a function of altitude to at least 35 km, and preferably higher in order to acquire data in a region of photochemical equilibrium. Availability of such data, really an extension of the ongoing measurements described by, for example, Evans et al.,[51] is an absolute prerequisite to establish definitively the role of aircraft NO_x emissions in regard to stratospheric ozone.

As discussed in the section on transport, we see a real absence of data that may be used to emulate or parameterize transport in the critical altitude region of 10 to 20 km or so. Discrepancies in model results due only to transport, such as those shown on Fig. 3, may turn out to be a fundamental limitation of our ability to predict effects of aircraft on ozone. If Fig. 3 were to represent the "final" state of things (which we suspect it does not), the difference in the calculated effect on ozone due only to use of two different transport parameterizations would be intolerable. It may mean the difference between predicting adverse and insignificant effects on ozone. Presently, there is no rigorously defensible means of determining which of the two parameterizations shown (of the many available) is "more valid."

Water-based constituents (the HO_x "family") play an important role in the stratospheric ozone balance. These constituents also directly and indirectly bear upon the sensitivity of stratospheric ozone amounts to changes in stratospheric concentrations of other species, including ClO_x and NO_x. Finally, they indirectly couple ClO_x and NO_x ozone cycles. Yet we do not have a reasonably good basis for assigning a value to the amount of water vapor in the stratosphere. The recent comprehensive review of Harries[52] demonstrates the point quite adequately. There are, then, two critical needs with regard to water vapor measurements: development of instruments capable of measuring water to heights of at least the stratopause (about 50 km), and accomplishment of the measurements themselves to provide a reasonable global stratospheric inventory of water. As a minimum initial step, lower stratospheric water measurements initiated by Mastenbrook[53] must be continued and expanded to several additional sites.

Although not discussed elsewhere in this paper, it is necessary to point out that the uncertainties in determination of aircraft exhaust NO_x concentrations first pointed out by McGregor et al.[54] have yet to be resolved. From our extensive discussions about the validity of their data, we can only conclude that one is neither able to dismiss it as inaccurate, nor, conversely, can one show the shortcomings of the technique (if there are any). Accordingly, one must acknowledge an uncertainty in the NO_x emission rates assigned to aircraft cruise operations. This uncertainty likely ranges somewhere between the order of tens of percent for "dry" engines and several times (up to a factor of 4 or 5) for engines that employ afterburning during cruise. Without knowing the reason for the discrepancy, one cannot be more precise.

As stated several times herein, the recent work of Howard and Evenson[22] carries major implications with regard to our understanding of the maintenance of the stratospheric ozone balance. This work must be confirmed by an independent experiment. In addition, the work of Duewer et al.[24] points up the crucial role played by a number of reactions in which the HO_x species participate. These also need to be measured (some have not) and/or independently verified.

We do not presently have a reasonable idea of the global ozone inventory or its variability with altitude, season, and latitude. This is a critical void and should be rectified as soon as possible. NASA is in the process of attempting to analyze data acquired since 1970 on its NIMBUS 4 satellite, but the complexity of such a task ("after the fact") makes the date of availability of this information uncertain. FAA's HAPP is sponsoring a study of the feasibility of acquiring global total ozone data from the Department of Defense Block 5D Satellite Series; ozone data from this study are not assured, because of the exploratory nature of the effort. We turn, then, to the "Ozone Data for the World," published under the auspices of the World Meteorological Organization.[55] But, as the spirited discussions at the Scientific Seminar on Stratospheric Monitoring (Paris, France, March 1977) showed, there are a number of problems in actually trying to use these data, *all* of which are always considered "preliminary." Clearly, the lack of validated, carefully analyzed data on the very substance at the center of all stratospheric studies is a situation that cannot be tolerated if the subject is to be taken seriously.

In the section on chemistry, we alluded to the complexities of the stratosphere's chemical system, the number of circuitous feedback paths, and the nonlinear nature of the system. We also later made reference to the complexities of stratospheric transport; Scorer[56] has pointed out the "layering" that can take place in this regard. In our view, there is a deep-seated need for exploration of the effects of these factors in models. Instead of being satisfied with accepting the results of models that compute results based on smoothly varying temperature and concentration profiles for all species, we must determine the sensitivity of these calculations to the jagged profiles that we know from actual measurement to be more realistic. It could turn out that nonlinearities of the stratospheric system will produce surprising results from such an analysis.

Finally, we note here the need to continue to use and improve upon the uncertainty analyses described earlier. It would be helpful if an analytic technique could be developed which could be economically employed to determine the overall sensitivity of a model output to changes in a given input parameter, especially if all other identifiable uncertainties were properly considered in the process. That is, although it is helpful to know the sensitivity of the output to a change in one input, all others being held fixed, this is not the complete story. The nonlinearity of these models could be such as to make a model appear to be insensitive to variations in one parameter, but if that parameter and another are varied simultaneously, the results might be vastly different. With as many uncertainties in input parameters as have already been discussed, it should not be surprising that we call for development of such an analytic capability.

Most of the foregoing research needs are presently being addressed, in one form or another. Major active programs in the United States include those of the Environmental Protection Agency (with regard to engine emission-rate measurement and fluorocarbon chemistry), the National Aeronautics and Space Administration (having a major research effort in this field), National Oceanic and Atmospheric Administration (especially through its Boulder Laboratories, the National Weather Service, and its Geophysical Fluid Dynamics Laboratory), the National Science Foundation (through its support of academic atmospheric sciences research generally and of the National Center for Atmospheric Research), and the Federal Aviation Administration (through its High Altitude Pollution Program). On an international level, there is considerable work in France and the United Kingdom, of which we are kept abreast through arrangements under the Tripartite Agreement on Stratospheric Monitoring,[14] and a number of specific bilateral agreements. The recent decision of the International Civil Aviation Organization to form a Committee on Aircraft Emissions chartered, among other things, to make recommendations on stratospheric emission standards, is a good example of the seriousness of purpose which the aviation community feels in regard to these questions. Lastly, we must point to the developing programs of the United Nations Environment Programme (UNEP) and the World Meteorological Organization in the field of stratospheric ozone. It is strongly hoped that UNEP will be successful in catalyzing the international scientific cooperation, especially with regard to atmospheric monitoring, which is so clearly necessary for proper understanding of these matters.

Concluding Remarks

From the preceding discussion, it should be evident that a number of significant changes in our understanding have taken place since the completion of the CIAP, NAS, COMESA, and COVOS studies. [A detailed summary of our understanding at the time of these studies (August 1975) may be found in Ref. 57.] The existence of these new data should not be allowed to create an atmosphere of complacency, however. Much of the information just described, especially with regard to new chemical reaction-rate data, needs independent verification before it can be accepted with confidence. Furthermore, in the previous section we outlined some of the major gaps that still exist in our understanding. Future data may not be as encouraging (from the aviation community's viewpoint) as has been the case with data acquired in the recent past.

We also hasten to point out that this discussion has focused on NO_x effects on ozone. What about those of water? The natural ozone production-destruction system must remain balanced, and the fact that ozone is less sensitive to NO_x increases than previously believed means that it must be more sensitive to other perturbations. ClO_x has taken up some of the "slack," and HO_x has absorbed the rest. Thus, detailed evaluation of perturbations to ozone from water in aircraft exhaust needs to be considered once again. Preliminary analysis of the sensitivity of ozone change to aircraft emissions of water, taking into account the most updated chemistry and radiative feedbacks, indicates[41] only a modest effect.

With the limitations that we have described here in mind, and drawing attention to the comprehensive summary of effects prepared by Oliver et al.,[15] we note the conclusions that we believe may be drawn:

1) Realistic projections to 1990 of air traffic growth for the world *subsonic* fleet of aircraft indicate that, given our present understanding, ozone will not be significantly affected by those aircraft, with a marginal increase in ozone now actually being predicted.

2) Furthermore, analysis of the effects of *subsonic* aircraft emissions (i.e., those below a nominal 15-km alt.) indicates, contrary to previous studies, that given our present understanding, subsonic aircraft NO_x emissions cannot foreseeably result in ozone reduction irrespective of fleet size (within reasonable bounds).

3) Earlier estimates[6-9] of ozone reduction for *SST* emissions of NO_x at 17 to 20 km appear to have been substantially in error. Based on simple 1-D models, our present understanding leads to predicted ozone perturbations of an absolute magnitude markedly smaller than given previously in these studies and outside their ranges of estimated uncertainty (if any were given).

4) For the first time, we can state that is is questionable whether NO_x emission from *"Concorde-like" SST's* (flying at about 17 m) reduce ozone at all. Present results, of 1-D models that consider both tropospheric and stratospheric chemistry, indicate that they do not, although these new data need verification before we can completely accept this important finding.

5) Similarly, the effect of NO_x emissions from aircraft cruising at 20 km is in doubt, although small ozone reductions for very large fleets of such aircraft are still predicted by some models.

6) Two-dimensional models (and 3-D models, if possible) must be used to corroborate the present results from 1-D models regarding SST perturbations; the ability of 1-D models to predict the direction of ozone perturbations (i.e., increase or decrease) is severely strained when the absolute magnitude of the effect is near zero, and latitudinal variations in average zone effects may be of importance in a complete assessment of the problem.

7) There is a clear need for more data on trace constituents in the atmosphere (especially simultaneous measurements of NO_x species, and measurements of HO_x species), for laboratory measurements of critical chemical reaction rates, for development and use of uncertainty analysis tools, and for a better understanding of transport in the 10- to 20-km-altitude region.

In summary, present understanding indicates that there is no imminent threat of ozone reduction from any type of existing aircraft, although substantial uncertainties remain. Thus there appears to be no immediate requirement to develop regulations that would mandate a reduction in cruise-altitude emissions of subsonic or supersonic aircraft. There is a clear necessity for continued study to reduce remaining uncertainties and to establish a uniform and internationally agreed accurate NO_x emission measurement scheme. Building a framework for regulation (if necessary) of future types of high-altitude aircraft, such as those frequently referred to as a "second-generation SST," or the even higher-flying hypersonic transport (HST), is also necessary. Activities of the new ICAO Committee on Aircraft Emissions should be encouraged to proceed in an orderly fashion along these lines.

Acknowledgments

We are indebted to a number of people for conversations on these matters, but especially thank J. Chang, P. Crutzen, E. Danielsen, W. Duewer, J. Mahlman, R. Oliver, N. Sundararaman, and D. Wuebbles for the time they have spent with us. This work was supported by the High Altitude Pollution Program of the Office of Environmental Quality, Federal Aviation Administration, U.S. Department of Transportation. The opinions expressed herein are solely those of the author and are not intended to reflect any policy of the Federal Aviation Administration or the U.S. Department of Transportation.

References

[1] McDonald, J., "Presentation before the Department of Commerce Technical Advisory Board on Environmental Aspects of the Supersonic Transport," Boulder, Colo., March 17-18, 1971.

[2] Harrison, H., "Stratospheric Ozone with Added Water Vapor: Influence of High-Altitude Aircraft," *Science*, Vol. 170, 1970, pp. 734-736.

[3] *Man's Impact on the Global Environment*, Report of the Study of Critical Environmental Programs (SCEP), Massachusetts Inst. of Technology Press, Cambridge, Mass., 1970.

[4] Crutzen, P.J., "The Influence of Nitrogen Oxides on the Atmospheric Ozone Content," *Quarterly Journal of the Royal Meteorological Society*, Vol. 96, 1970, pp. 320-325; also Crutzen, P.J., "SST's—A Threat to the Earth's Ozone Shield?," *Ambio*, Vol. 1, 1972, pp. 41-51.

[5] Johnston, H.S., "Reduction of Stratospheric Ozone by Nitrogen Oxide Catalysts from SST Exhaust," *Science*, Vol. 173, 1971, pp. 517-522.

[6] Grobecker, A.J., Coroniti, S.C., and Cannon, R.H. Jr., "CIAP Report of Findings: The Effects of Stratospheric Pollution by Aircraft," U.S. Dept. of Transportation, Washington, D.C., Rept. DOT-TST-75-50, 1974.

[7] "Environmental Impact of Stratospheric Flight," National Academy of Sciences, Climatic Impact Committee, Washington, D.C., 1975.

[8] "The Report of the Committee on Meteorological Effects of Stratospheric Aircraft (COMESA), 1972-1975," U.K. Meteorological Office, Bracknell, England, 1975.

[9] "Comite d'Etudes sur les Consequences des Vols Stratospheriques (COVOS), Activites 1972-1976," Societe Meteorologique de France, Boulogne, France, 1976.

[10] Smykov, V.G., "TU-144 Emissions," *Report of the Fourth Meeting of the Technical Panel on Supersonic Transport Operations*, International Civil Aviation Organization, ICAO Doc. 9076, 1973.

[11] Rummel, R.W., "Possible Impact of Regulations on Airlines," *Proceedings of the Fourth Conference on the Climatic Impact Assessment Program*, U.S. Dept. of Transportation, Washington, D.C., Rept. DOT-TSC-OST-75-38, 1976, pp. 127-131.

[12] Meister, F.A., "Panel Discussion," *Proceedings of the Fourth Conference on the Climatic Impact Assessment Program*, U.S. Dept. of Transportation, Washington, D.C., Rept. DOT-TSC-OST-75-38, 1976, pp. 24-38.

[13] "The Secretary's Decision on Concorde," U.S. Dept. of Transportation, Washington, D.C., Feb. 4, 1976.

[14] "Tripartite Agreement on Stratospheric Monitoring," concluded among France, the United Kingdom, and the United States, May 5, 1976.

[15] Oliver, R.C., Bauer, E., Hidalgo, H., Gardner, K.A., and Wasylkiwskyj, W., "Aircraft Emissions: Potential Effects on Ozone and Climate," U.S. Dept. of Transportation, Washington, D.C., Rept. FAA-EQ-77-3, March 1977.

[16] Chapman, S., "A Theory of Upper-Atmosphere Ozone," *Memoirs, Royal Meteorological Society*, Vol. 3, 1930, pp. 103-125.

[17] Johnston, H.S., "Global Ozone Balance in the Natural Stratosphere," *Reviews of Geophysics and Space Physics*, Vol. 13, 1975, pp. 637-649.

[18] Hampson, J., "Photochemical Behavior of the Ozone Layer," Canadian Armament Research and Development Establishment, (CARDE), Valcartier, Quebec, Canada, TN 1627/64, 1964.

[19] Hunt, B.G., "Photochemistry of Ozone in a Moist Atmosphere," *Journal of Geophysical Research*, Vol. 71, 1966, pp. 1385-1398.

[20] Stolarski, R.S., and Cicerone, R.J., "Stratospheric Chlorine: Possible Sink for Ozone," *Canadian Journal of Chemistry*, Vol. 52, 1974, pp. 1610-1615.

[21] "Halocarbons: Effects on Stratospheric Ozone," quotation on Space Shuttle estimate, National Academy of Sciences, Panel on Atmospheric Chemistry, Washington, D.C., 1976, pp. 164-165.

[22] Howard, C.H. and Evenson, K.M., "Laser Magnetic Resonance Study of HO_2 Chemistry," *AGU Annual Spring Meeting*, Washington, D.C., May/June 1977.

[23] "Halocarbons: Effects on Stratospheric Ozone," National Academy of Sciences, Panel on Atmospheric Chemistry, Washington, D.C., 1976.

[24] Duewer, W.H., Wuebbles, D.J., Ellsaesser, H.W., and Chang, J.S., "NO_x Catalytic Ozone Destruction: Sensitivity to Rate Coefficients," *Journal of Geophysical Research*, Vol. 82, 1977, pp. 935-942.

[25] Johnston, H.S. and Nelson, J., "Comment on 'NO_x Catalytic Ozone Destruction: Sensitivity to Rate Coefficients' by W.H. Duewer et al.," *Journal of Geophysical Research*, Vol. 82, 1977, pp. 2593-2598.

[26] Duewer, W.H., Wuebbles, D.J., Ellsaesser, H.W., and, Chang, J.S., "Reply," *Journal of Geophysical Research*, Vol. 82, 1977, pp. 2599-2605.

[27] Hampson, R.F., Jr. and Garvin, D., "Chemical Kinetic and Photochemical Data for Modelling Atmospheric Chemistry," National Bureau of Standards, TN 866, June 1975.

[28] Smagorinsky, J., "Global Atmospheric Modeling and Numerical Simulation of Climate," *Weather and Climate Modification*, edited by W.N. Hess, Wiley, New York, 1974.

[29] "Understanding Climatic Change," National Academy of Science, U.S. Committee for the Global Atmospheric Research Program, Washington, D.C., 1975.

[30] Mahlman, J.D., "Some Fundamental Limitations of Simplified-Transport Models as Implied by Results from a Three-Dimensional General-Circulation/Tracer Model," *Proceedings of the Fourth Conference on the Climatic Impact Assessment Program*, U.S. Dept. of Transportation, Washington, D.C., Rept. DOT-TSC-OST-75-38, 1976, pp. 132-146.

[31] Mahlman, J.D., private communication, 1977.

[32] Cunnold, D., Alyea, F., Phillips, N., and Prinn, R., "A Three-Dimensional Dynamical-Chemical Model of Atmospheric Ozone," *Journal of the Atmospheric Sciences*, Vol. 32, 1975, pp. 170-194.

[33] Widhopf, G., Glatt, L., and Kramer, R., "A Two-Dimensional Time-Dependent Photochemical Model of the Atmosphere," Aerospace Corp., Final Rept., Oct. 1976.

[34] Hidalgo, H. and Crutzen, P.J., "The Tropospheric and Stratospheric Composition Perturbed by NO_x Emissions of High-Altitude Aircraft," *Non-Urban Tropospheric Composition Symposium*, Miami, Fla., Nov. 9-12, 1976.

[35] Reed, R.J., "A Study of a Characteristic Type of Upper Level Frontogenesis," *Journal of Meteorology*, Vol. 12, 1955, pp. 226-237.

[36] Reed, R. and Danielsen, E.F., "Fronts in the Vicinity of the Tropopause," *Archiv für Meteorologie, Geophysik und Bioklimatologie*, Ser. A., Vol. 2, 1959, p. 1.

[37] Danielsen, E.F., "Transport from Stratosphere to Troposphere," *Mineral Industry*, Vol. 33, 1964.

[38] Danielsen, E.F., "Stratospheric-Tropospheric Exchange Based on Radioactivity, Ozone and Potential Vorticity," *Journal of the Atmospheric Sciences*, Vol. 25, 1968, pp. 502-518.

[39] Chang, J.S., "Coupled Chemical Kinetics and Atmospheric Transport Models of the Stratosphere," *173rd Meeting of the American Chemical Society*, Anaheim, Calif., March 1977.

[40] Wuebbles, D., private communication, 1976.

[41] Chang, J.S., private communication, 1977.

[42] Chang, J.S. and Johnston, H.S., "The Effect of NO_x Effluents on Ozone," *Proceedings of the Third Conference on the Climatic Impact Assessment Program*, U.S. Dept. of Transportation, Washington, D.C., Rept. DOT-TSC-OST-74-15, 1974, pp. 323-329 (model referred to is "model 4").

[43] Bates, D.R. and Hays, P.B., "Atmospheric Nitrous Oxide," *Planetary and Space Science*, Vol. 15, 1967, p. 189.

[44] Johnston, H.S. and Selwyn, G.S., "New Cross Sections for the Absorption of Near-Ultraviolet Radiation by Nitrous Oxide (N_2O)," *Geophysical Research Letters*, Vol. 2, 1975, p. 549.

[45] Streit, G.E., Howard, C.J., Schmeltekopf, A.L., Davidson, J.A., and Schiff, M.I., "Temperature Dependence of $O(^1D)$ Rate Constants for Reactions with O_2, N_2, CO_2, O_3 and H_2O," *The Journal of Chemical Physics*, Vol. 65, 1976, pp. 4761-4764.

[46] Tsang, W., "Comparisons Between Experimental and Calculated Rate Constants for Dissociation and Combination Reactions Involving Small Polyatomic Molecules," *International Journal of Chemical Kinetics*, Vol. 5, 1973, p. 947.

[47] Anastasi, C. and Smith, I.W.M., "Rate Measurements of Reactions of OH by Resonance Absorption," *Journal of the Chemical Society, Faraday Transactions II*, Vol. 72, 1976, p. 1459.

[48] Hunten, D.M., "The Philosophy of One-Dimensional Modeling," *Proceedings of the Fourth Conference on the Climatic Impact Assessment Program*, U.S. Dept. of Transportation, Washington, D.C., Rept. DOT-TSC-OST-75-38, 1976, pp. 147-155.

[49] Stolarski, R.S., Butler, D.M., and Rundel, R.D., "Uncertainty in Stratospheric Ozone Perturbation Predictions Due to Uncertainties in Reaction Rates," *International Conference on Problems Related to the Stratosphere*, Logan, Utah, Sept. 15-17, 1976.

[50] Ackerman, M., Frimout, D., and Muller, C., "Stratospheric Methane—Measurements and Predictions," Aeronomica Acta, A-No. 180, 1977.

[51] Evans, W.F.J., Kerr, J.B., and Ridley, B.A., "Measurements of the Nitrogen Chemistry of the Ozone Layer from Project

Stratoprobe," *International Conference on Problems Related to the Stratosphere,* Logan, Utah, Sept. 15-17, 1976.

[52] Harries, J.E., "The Distribution of Water Vapor in the Stratosphere," *Reviews of Geophysics and Space Physics,* Vol. 14, 1976, pp. 565-575.

[53] Mastenbrook, H.J., "Water Vapor Distribution in the Stratosphere and High Troposphere," *Journal of the Atmospheric Sciences,* Vol. 25, 1968, pp. 299-311.

[54] McGregor, W.K., Seiber, B.L., and Few, J.D., "Concentration of OH and HO in YJ93-GE-3 Engine Exhausts Measured In Situ by Narrow-Line UV Absorption," *Proceedings of the Second Conference on the Climatic Impact Assessment Program,* U.S. Dept. of Transportation, Washington, D.C., Rept. DOT-TSC-OST-73-4, 1973, pp. 214-229.

[55] "Ozone Data for the World," Meteorological Branch, Canadian Dept. of Transportation, Toronto, Canada, 1960—.

[56] Scorer, R.S., "A Commentary on Ozone Depletion Theories," *Atmospheric Environment,* Vol. 10, 1976, pp. 177-180.

[57] Broderick, A.J., "Effects of Cruise-Altitude Pollution," *Journal of Aircraft,* Vol. 13, Oct. 1976, pp. 817-822.

From the AIAA Progress in Astronautics and Aeronautics Series . . .

STRATOSPHERIC CIRCULATION—v. 22

Edited by Willis L. Webb, White Sands Missile Range, and University of Texas at El Paso

Thirty papers in this volume present data from synoptic rocket exploration of the stratosphere, disclosing fundamental circulation, and tidal and global circulation systems.

A summary of stratospheric circulation is presented, detailing the state of knowledge and the Meteorological Rocket Network (MRN) and the development of the Stratospheric Circulation Index (SCI), with the outlook for a global sounding rocket network.

Covers the British Skua rocket system, the Polish Meteor-I, the Air Force PWN-8B (Loki-Dart) and ARCAS meteorological rockets, and the Kookaburra dropsonde rocket system.

Examinations of meteorological sensing systems cover radar chaff, parachute-borne sensors, balloon sounding, resistance wire and thermistor use in temperature sensing, and ozonesondes.

Atmosphere studies cover clouds, mesospheric ozone, and a photochemical model of the mesosphere. Upper atmosphere studies cover specific geographic areas and upper atmosphere climatology, comparing current data with previous studies and the Standard Atmosphere Supplements.

600 pp., 6 x 9, illus. $18.00 Mem. & List

TO ORDER WRITE: Publications Dept., AIAA, 1290 Avenue of the Americas, New York, N. Y. 10019