

CHAPTER 11

Subsonic and Supersonic Aircraft Emissions

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CHAPTER 11

SUBSONIC AND SUPERSONIC AIRCRAFT EMISSIONS

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SCIENTIFIC SUMMARY

Extensive research and evaluations are underway to assess the atmospheric effects of the present and future subsonic aircraft fleet and of a projected fleet of supersonic transports. Assessment of aircraft effects on the atmosphere involves the following:

- i) measuring the characteristics of aircraft engine emissions;
- ii) developing three-dimensional (3-D) inventories for emissions as a function of time;
- iii) developing plume models to assess the transformations of the aircraft engine emissions to the point where they are governed by ambient atmospheric conditions;
- iv) developing atmospheric models to assess aircraft influences on atmospheric composition and climate; and
- v) measuring atmospheric trace species and meteorology to test the understanding of photochemistry and transport as well as to test model behavior against that of the atmosphere.

Supersonic and subsonic aircraft fly in atmospheric regions that have quite different dynamical and chemical regimes. Subsonic aircraft fly in the upper troposphere and in the stratosphere near the tropopause, where stratospheric residence times due to exchange with the troposphere are measured in months. Proposed supersonic aircraft will fly in the stratosphere near 20 km, where stratospheric residence times due to exchange with the troposphere increase to years. In the upper troposphere, increases in NO_x typically lead to increases in ozone. In the stratosphere, ozone changes depend on the complex coupling among HO_x , NO_x , and halogen reactions.

- Emission inventories have been developed for the current subsonic and projected supersonic and subsonic aircraft fleets. These provide reasonable bases for inputs to models. Subsonic aircraft flying in the North Atlantic flight corridor emit 56% of their exhaust emissions into the upper troposphere and 44% into the lower stratosphere on an annual basis.
- Plume processing models contain complex chemistry, microphysics, and turbulence parameterizations. Only a few measurements exist to compare to plume processing model results.
- Estimates indicate that present subsonic aircraft operations may have increased NO_x concentrations at upper tropospheric altitudes in the North Atlantic flight corridor by about 10-100%, water vapor concentrations by about 0.1% or less, SO_x by about 10% or less, and soot by about 10% compared with the atmosphere in the absence of aircraft and assuming all aircraft are flying below the tropopause.
- Preliminary model results indicate that the current subsonic fleet produces upper tropospheric ozone increases as much as several percent, maximizing at the latitudes of the North Atlantic flight corridor.
- The results of these rather complex models depend critically on NO_x chemistry. Since there are large uncertainties in the present knowledge of the tropospheric NO_x budget (especially in the upper troposphere), little confidence should be put in these quantitative model results of subsonic aircraft effects on the atmosphere.
- Atmospheric effects of supersonic aircraft depend on the number of aircraft, the altitude of operation, the exhaust emissions, and the background chlorine and aerosol loading. Rough estimates of the impact of future supersonic operations (assuming 500 aircraft flying at Mach 2.4 in the stratosphere and emitting 15 grams of nitrogen oxides per kilogram of fuel) indicate an increase of the North Atlantic flight corridor concentrations of NO_x up to about 250%, water vapor up to about 40%, SO_x up to about 40%, H_2SO_4 up to about 200%, soot up to about 100%, and CO up to about 20%.

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- One result of two-dimensional model calculations of the impact of such a projected fleet in a stratosphere with a chlorine loading of 3.7 ppbv (corresponding to the year 2015) implies additional annually averaged ozone column decreases of 0.3-1.8% for the Northern Hemisphere. Although NO_x aircraft emissions have the largest impact on ozone, the effects from H_2O emissions contribute to the calculated ozone change (about 20%).
- Net changes in the column ozone from supersonic aircraft modeling result from ozone mixing ratio enhancements in the upper troposphere and lower stratosphere and depletion at higher stratospheric altitudes.
- There are important uncertainties in supersonic assessments. In particular, these assessment models produce ozone changes that differ among each other, especially in the lower stratosphere below 25 km. When used to calculate ozone trends, these same models predict smaller changes than are observed in the stratosphere below 25 km between 1980 and 1990. Thus, these models may not be properly including mechanisms that are important in this crucial altitude range.
- Increases in ozone at altitudes near the tropopause, such as are thought to result from aircraft emissions, enhance the atmosphere's greenhouse effect. Research to evaluate the climate effects of supersonic and subsonic aircraft operations is just beginning, so reliable quantitative results are not yet available, but some initial estimates indicate that this effect is of the same order as that resulting from the aircraft CO_2 emissions.

11.1 INTRODUCTION

Tremendous growth occurred in the aircraft industry during the last several decades. Figure 11-1 shows the increasing use of aircraft fuel as a function of time. Aircraft fuel consumption has increased by about 75% during the past 20 years and is projected to increase by 100 to 200% over the next 30 years. At the present time, approximately 3% of the worldwide usage of fossil fuels is by aircraft. Ninety-nine percent of this aircraft fuel is burned by subsonic aircraft, of which a large proportion occurs in the upper troposphere. Table 9.2 of the previous assessment (WMO, 1992) demonstrates that subsonic aircraft emit a significant fraction of their exhaust products into the lower stratosphere. This depends on factors such as latitude and season.

Despite the small percentage of the total fossil fuel usage for aviation, the environmental effects of aircraft should be closely examined for several reasons. One reason is the rapid growth that has occurred and is projected to occur in aircraft emissions, and another is that aircraft emit their exhaust products at specific altitudes where significant effects might be expected. For instance, an environmental concern of the 1970s was the effect that large fleets of supersonic aircraft would have on the stratospheric ozone layer. The main concern was then and still is that catalytic cycles involving aircraft-emitted NO_x (NO plus NO_2) enhance the destruction of ozone.

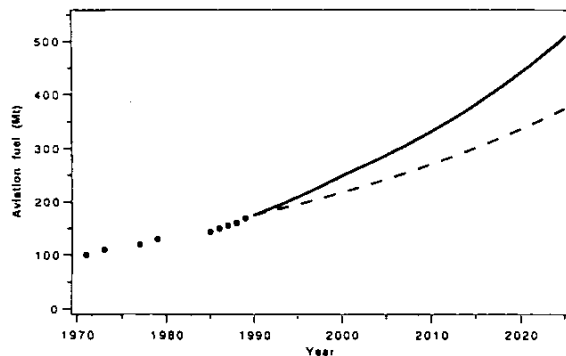


Figure 11-1. Aviation fuel versus time. Data up to 1989 from the International Energy Agency (1990). Extrapolations according Kavanaugh (1988) with 2.2% per year in a low-fuel scenario and with 3.6% up to 2000 and 2.9% thereafter in a high-fuel scenario. (Based on Schumann, 1994.)

Since supersonic aircraft engines may emit significant amounts of NO_x , the fear is that large fleets of supersonic aircraft flying at stratospheric levels, where maximum ozone concentrations exist, might seriously deplete the stratospheric ozone layer, leading to increased ultraviolet radiation flux on the biosphere. Also, climate sensitivity studies have shown that ozone changes in the upper troposphere and lower stratosphere will have greater radiative effects on changing surface and lower tropospheric temperatures than would ozone changes at other levels (see Chapter 8).

Also, in the 1950s, "smog reactions" were discovered that implied the depletion of tropospheric ozone when NO_x concentrations are low and ozone production when NO_x concentrations are high. Thus, there is a concern that new fleets of supersonic aircraft flying in the stratosphere would lead to harmful stratospheric ozone depletion, while present and future subsonic aircraft operations will lead to undesired enhanced levels of ozone in the upper troposphere.

Development of any successful aircraft requires a period of about 25 years, and each aircraft will have a useful lifetime of about 25 years as well. Thus, even if an environmentally motivated decision is made to utilize new aircraft technologies, it will take decades to fully realize the benefits.

One can get some perspective on possible atmospheric effects of aircraft operations by noting the following. Current subsonic aircraft operations in the North Atlantic flight corridor are probably increasing NO_x concentrations at upper tropospheric altitudes by about 10-100%, water vapor concentrations by about 0.1% or less, and SO_x by about 10% or less compared to an atmosphere without aircraft. Future supersonic operations in the stratosphere might increase the North Atlantic flight corridor concentrations of NO_x up to about 250%, water vapor up to about 40%, SO_x up to about 40%, H_2SO_4 up to about 200%, soot up to about 100%, and CO up to about 20%. Thus, present subsonic aircraft perturbations in atmospheric composition are now probably significant, and future large supersonic aircraft fleet operations will also be significant in affecting atmospheric trace gas concentrations.

These and other concerns have led to an increasing amount of research into the atmospheric effects of current and future aircraft operations. In the U.S., NASA's Atmospheric Effects of Aviation Project is composed of

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two elements. The Atmospheric Effects of Stratospheric Aircraft (AESA) element was initiated in 1990 to evaluate the possible impact of a proposed fleet of high-speed (*i.e.*, supersonic) civil transport (HSCT) aircraft. A Subsonic Assessment (Wesoky *et al.*, 1994) was begun in 1994 to study the impact of the current commercial aircraft fleet. In Europe, the Commission of the European Communities (CEC) has initiated the Impact of NO_x Emissions from Aircraft upon the Atmosphere (AERONOX) and Measurement of Ozone on Airbus In-service Aircraft (MOZAIC) programs (Aeronautics, 1993) and Pollution from Aircraft Emissions in the North Atlantic Flight Corridor (POLINAT) to investigate effects of the emissions of the present subsonic aircraft fleet in flight traffic corridors. In addition, there are also several national programs in Europe and Japan looking at various aspects of the atmospheric effects of aircraft emissions.

Atmospheric models play a particularly important role in these programs since there does not appear to be any purely experimental approach that can evaluate the global impact of aircraft operations on the atmosphere. The strategy is to construct models of the present atmosphere that compare well with atmospheric measurements and to use these models to try to predict the future atmospheric effects of changed aircraft operations. At the present time, the subsonic and supersonic assessment programs are in quite different stages of maturity and are utilizing different approaches in both modeling and observations. Therefore, in this chapter the subsonic and supersonic evaluations will be considered separately since the chemical and dynamical regimes are quite different. In this context the "lower stratosphere" refers to the region above the local tropopause where there are lines of constant potential temperature that connect the stratosphere and troposphere. In this region, stratosphere-troposphere exchange can occur by horizontal advection with no need to expend energy in overcoming the stable stratification. In the stratosphere near 20 km, where Mach 2.4 HSCT operate, no lines of constant potential temperature connecting the stratosphere and troposphere exist. Therefore residence times of tracers are much larger (about 2 years) in the stratosphere at 20 km than in the lower stratosphere.

In this chapter, we will review what is known about aircraft emissions into the atmosphere and discuss the transformations that take place in the aircraft plume

as it adjusts from the physical conditions of the aircraft exhaust leaving the engine tailpipe to those of the ambient atmosphere. Some of the atmospheric effects of the different chemical families that are emitted by aircraft are then considered, and finally, modeling studies of the atmospheric effects of aircraft emissions on ozone are presented, along with a discussion of possible climate effects of aircraft operations. A discussion of the level of uncertainty of these predictions, and some conclusions are presented.

Further details of the NASA effort to assess the atmospheric effects of future supersonic aircraft operations can be found in Albritton *et al.* (1993) and the references therein. An external evaluation of these efforts can be found in NRC (1994). No similar documents exist at this time pertaining to the atmospheric effects of subsonic aircraft operations.

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The evaluation of the potential impact of emissions from aircraft on atmospheric ozone levels requires a comprehensive understanding of the nature of the emissions produced by all types of aircraft and a knowledge of the operations of the total global aircraft fleet in order to generate a time-dependent, three-dimensional emissions data base for use in chemical/dynamical atmospheric models.

Emissions from the engines, rather than those associated with the airframe, are considered to be dominant (Prather *et al.*, 1992). These are functions of engine technology and the operation of the aircraft on which the engines are installed. Primary engine exhaust products are CO₂ and H₂O, which are directly related to the burned fuel, with minor variations due to the precise carbon-hydrogen ratio of the fuel. Secondary products include NO_x (= NO + NO₂), CO, unburned and partially burnt fuel hydrocarbons (HC), soot particulates/smoke, and SO_x. NO_x is a consequence of the high temperature in the engine combustor; the incomplete combustion products (CO, HC, and soot/smoke) are functions of the engine design and operation and may vary widely between engines. SO_x is directly related to fuel composition. Currently, typical sulfur levels in aviation kerosene are about 0.05% sulfur by weight, compared with an allowed specification limit of 0.3% (ICAO, 1993).

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Table 11-1. Emission Index (grams per kilograms of fuel used) of various materials for subsonic and supersonic aircraft for cruise condition. Values in parentheses are ranges for different engines and operating conditions.

| Species (gm MW) | Subsonic Aircraft* | | Supersonic Aircraft* |
|---|--------------------|-----------------|-----------------------------|
| | Short range | Long range | |
| CO ₂ (44) | 3160 | 3160 | 3160 |
| H ₂ O (18) | 1230 | 1230 | 1230 |
| CO (28) | 5.9 (0.2-14) | 3.3 (0.2-14) | 1.5 (1.2-3.0) |
| HC as methane (16) | 0.9 (0.12-4.6) | 0.56 (0.12-4.6) | 0.2 (0.02-0.5) |
| SO ₂ (64) | 1.1 | 1.1 | 1.0 |
| NO _x as NO ₂ (46) | 9.3 (6-19) | 14.4 (6-19) | depends on design (5-45) |

* Mean (fuel-consumption weighted) emission indices for 1987 based on Boeing (1990). The values were calculated from a data base containing emission indices and fuel consumptions by aircraft types. The difference between short range (cruise altitude around 8 km) and long range (cruise altitude between 10 and 11 km) reflects different mixes of aircraft used for different flights.

Based on Boeing (1990) and McDonnell Douglas (1990).

The measure of aircraft emissions traditionally used in the aviation community is the Emissions Index (EI), with units of grams per kilogram of burnt fuel. Typical EI values for subsonic and anticipated values for supersonic aircraft engines are given in Table 11-1 for cruise conditions. By convention, EI(NO_x) is defined in terms of NO₂ (similarly, hydrocarbons are referenced to methane).

Historically, the emissions emphasis has been on limiting NO_x, CO, HC, and smoke, mainly for reasons relating to boundary layer pollution. Standards are in place for control of these over a Landing/Take-Off (LTO) cycle up to 915 m altitude at and around airports (ICAO, 1993). Currently there are no regulations covering other flight regimes, *e.g.* cruise, though ICAO (1991) is considering the need and feasibility of introducing standards.

It is now recognized that the list of chemical species (emitted from engines or possibly produced in the young plume, also by reactions with ambient trace species like hydrocarbons) that may be relevant to ozone and climate change extends well beyond the primary combustion species and NO_x. A more complete set of "odd nitrogen" compounds, known as NO_y—including NO_x, N₂O₅, NO₃, HNO₃—and PAN (peroxyacetylni-

trate) should be considered, along with SO_x and soot particles as aerosol-active species. HC and CO may also play an important role in high altitude HO_x chemistry.

11.2.1 Subsonic Aircraft

Engine design is a compromise between many conflicting requirements, among which are safety, economy, and environmental impacts. For subsonic engines, the various manufacturers have resolved these conflicts with different compromises according to their own in-house styles. This has resulted in a spread of emission values for HC, CO, NO_x, and smoke, all meeting the LTO cycle regulatory standards.

Historical trends (1970–1988) in aircraft engine emissions for the typical LTO cycle show that very substantial decreases in HC and CO emissions have been realized over the past two decades due to improvements in fuel-efficient engine design and emissions control technology. A substantial increase in NO_x emissions would have been expected due to the much higher combustion temperatures associated with the more fuel-efficient engine cycles. However, other improvements in engine technology have kept NO_x relatively constant. Combining the increased passenger miles in the period from 1970 to 1988 with that of the technology improvements

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would imply that the actual mass output should have decreased by about 77% for HC and 30% for CO, while NO_x mass output should have increased by about 110%. Considerable further reductions of HC and CO will come as older aircraft are phased out, but little change can be expected for NO_x without the introduction of low- NO_x technology engines.

The first steps to develop combustion systems producing significantly lower NO_x levels relative to existing technology were made in the mid-1970s (CIAP 2, 1975). These systems achieve at least a 30% NO_x reduction, and are now being developed into airworthy systems for introduction in medium and high thrust engines.

11.2.2 Supersonic Aircraft

The first generation of civil supersonic aircraft (Concorde, Tupolev TU144) incorporated turbojet engines of a technology level typical of the early 1970s. The second generation, currently being considered by a number of countries and industrial consortia, will have to incorporate technology capable of meeting environmental requirements. A comprehensive study of the scientific issues associated with the Atmospheric Effects of Stratospheric Aircraft (AESAs) was initiated in 1990 as part of NASA's High Speed Research Program (HSRP; Prather *et al.*, 1992). No engines or prototypes exist and designs are only at the concept stage. A range of cruise EI(NO_x) levels (45, 15, and 5) has been set as the basis for use in atmospheric model assessments and in developing engine technology. An EI(NO_x) of 45 is approximately what would be obtained if HSCT engines were to be built using today's jet engine technology without putting any emphasis on obtaining lower EI(NO_x) emissions. Jet engine experts have great confidence in their ability to achieve an HSCT engine design with EI(NO_x) no greater than 15 and have set a goal of designing an HSCT engine with EI(NO_x) no greater than 5. Laboratory-scale studies of new engine concepts, which appear to offer the potential of at least 70-80% reduction in NO_x compared with current technology, are being pursued. Early results indicate that these systems seem able to achieve the low target levels of EI(NO_x) = 5 (Albritton *et al.*, 1993).

11.2.3 Military Aircraft

In contrast to the majority of civil aviation, military aircraft do not operate to set flight profiles or

frequencies. Also, national authorities are reluctant to disclose this information. Thus it is extremely difficult to make realistic assessments of the contribution of military aircraft in terms of fuel usage or emissions. Earlier estimates (Wuebbles *et al.*, 1993) were that the world's military aircraft used about 19% of the total aviation fuel and emitted 13% of the NO_x , with an average EI(NO_x) of 7.5. With the changes following the breakup of the former Soviet Union, there has been considerable reduction in activity, and an estimate of about 10% fuel usage may be more appropriate (ECAC/ANCA, 1994).

11.2.4 Emissions at Altitude

As noted above, engines are currently only regulated for some species over an LTO cycle. Internationally accredited emissions data on these are available (ICAO, 1994). However, experimental data for other flight conditions are sparse, since these can only realistically be obtained from tests in flight or in altitude simulation test facilities. Correlations, in particular for NO_x , have been developed from theoretical studies and combustor test programs for prediction of emissions over a range of flight conditions. A review of these is given elsewhere (Prather *et al.*, 1992; Albritton *et al.*, 1993). Engine tests under simulated altitude conditions are being carried out within the AERONOX program (Aeronautics, 1993) and should be useful to check this approach for subsonic engines.

11.2.5 Scenarios and Emissions Data Bases

Air traffic scenarios have been developed as a basis for evaluating global distributions of emissions from aircraft (McInnes and Walker, 1992; Prather *et al.*, 1992; Wuebbles *et al.*, 1993; ECAC/ANCA, 1994). The first two based their traffic assessment on scheduled commercial flight information from timetables and supplemented these data with information from other sources for non-scheduled charter, general aviation, and military flights. The third is based on worldwide Air Traffic Control data supplemented by timetable information and other data as appropriate.

McInnes and Walker (1992) generated 2-D and 3-D inventories of NO_x emissions from subsonic aircraft, using relatively broad assumptions for numbers of aircraft types, flight profiles/distance bands, and cell sizes. However, the evaluation did not include non-scheduled, military, cargo, or general aviation, and

both inventories accounted for only 51% of the total estimated fuel consumption of 166.5×10^9 kg for the year 1989 (IEA, 1990). The fuel consumption was simply scaled to match the total estimated fuel consumption in order to estimate the total NO_x mass. Their average EI(NO_x) value of 11.6 is within the range quoted elsewhere (Nüßer and Schmitt [1990] 6 - 16.4; Egli [1990] 11-30; and Beck *et al.* [1992] 17.9).

Wuebbles *et al.* (1993) generated for the HSRP/AESA (Prather *et al.*, 1992; Stolarski and Wesoky, 1993a) a comprehensive assessment of all aircraft types to determine fuel, NO_x, CO, and HC for general scenarios comprising the 1990 fleet and projected fleets of subsonic and supersonic aircraft (HSCTs) for the year 2015. A much better match (76%) of the calculated fuel use with the total estimated fuel consumption for 1990 was achieved. The remainder is likely to be mainly attributable to factors such as the non-idealized flight routings and altitudes actually flown by aircraft due to factors such as air traffic control, adverse weather, etc., as well as low-level unplanned delays and ground operations. However, scaling to match the total estimated fuel consumption gave a total annual NO_x mass (1.92 Tg) similar to that of McInnes and Walker. Illustrations of the global NO_x inventories as functions of latitude/longitude, or altitude/latitude for both 1990 and 2015 are given in Figures 11-2 and 11-3.

The European Civil Aviation Conference (ECAC) Abatement of Nuisance Caused by Air Traffic (ANCAT) work, carried out to complement the AERONOX program, has also considered NO_x emissions from subsonic and supersonic fleets for the year 1992. Unlike the other inventories, the traffic data have been compiled for four equally spaced months throughout the year to provide information on the seasonal variation. Preliminary results indicate a higher fuel burn, NO_x annual mass, and

EI(NO_x) than those of the other inventories and are likely to represent upper bounds on the aircraft NO_x emission burden. The current grid scale is larger than that of the HSRP/AESA inventory, but this may give a more realistic representation of the NO_x distribution within the heavily traveled air traffic routes, such as the North Atlantic, where there is known to be a significant divergence of actual flight paths from the ideal great circle routes currently assumed by all inventories. Further work is being carried out to produce forecast inventories for the years 2003 and 2015.

Considerable comparative analysis is being undertaken between the ECAC/ANCAT and the HSRP/AESA inventories in order to understand the reasons underlying the differences (EI(NO_x) 10.9 to 16.8; NO_x mass 1.92 to 2.8 Tg) and to refine the inventories. For example, it is already known that there is some double counting of traffic in some geographically important areas of the ECAC/ANCAT inventory. Another significant factor is a large difference in the contribution from military aircraft. A comparison summary of the inventories is given in the table at the bottom of the page.

11.2.6 Emissions Above and Below the Tropopause

In a global perspective, the North Atlantic, apart from North America and Europe, contains the largest specific subsonic traffic load. In 1990 the average daily movements across the Atlantic (both directions) between 45° and 60°N amounted to 595 flights in July and 462 flights in November. One recent study (Hoinka *et al.*, 1993) has assessed the aircraft fleet mix and the resulting emissions for this flight corridor. By correlation of the traffic data with the tropopause height from the European Centre for Medium-Range Weather Forecasts

| | <i>McInnes and Walker,</i> 1992 | <i>Wuebbles et al.,</i> 1993 | <i>ECAC/ANCAT,</i> 1994 |
|------------------------------|------------------------------------|---------------------------------|----------------------------|
| Year | 1989 | 1990 | 1992 |
| Grid size | 7.5° × 7.5° × 0.5km | 1° × 1° × 1km | 2.8° × 2.8° × 1km |
| Fuel match | 51% | 76% | 99% |
| EI (NO _x) global | 11.6 | 10.9 | 16.8 |
| NO _x mass (Tg)# | 1.91# | 1.92# | 2.8# |

Note: all data for NO_x mass have been scaled to 100% fuel match.

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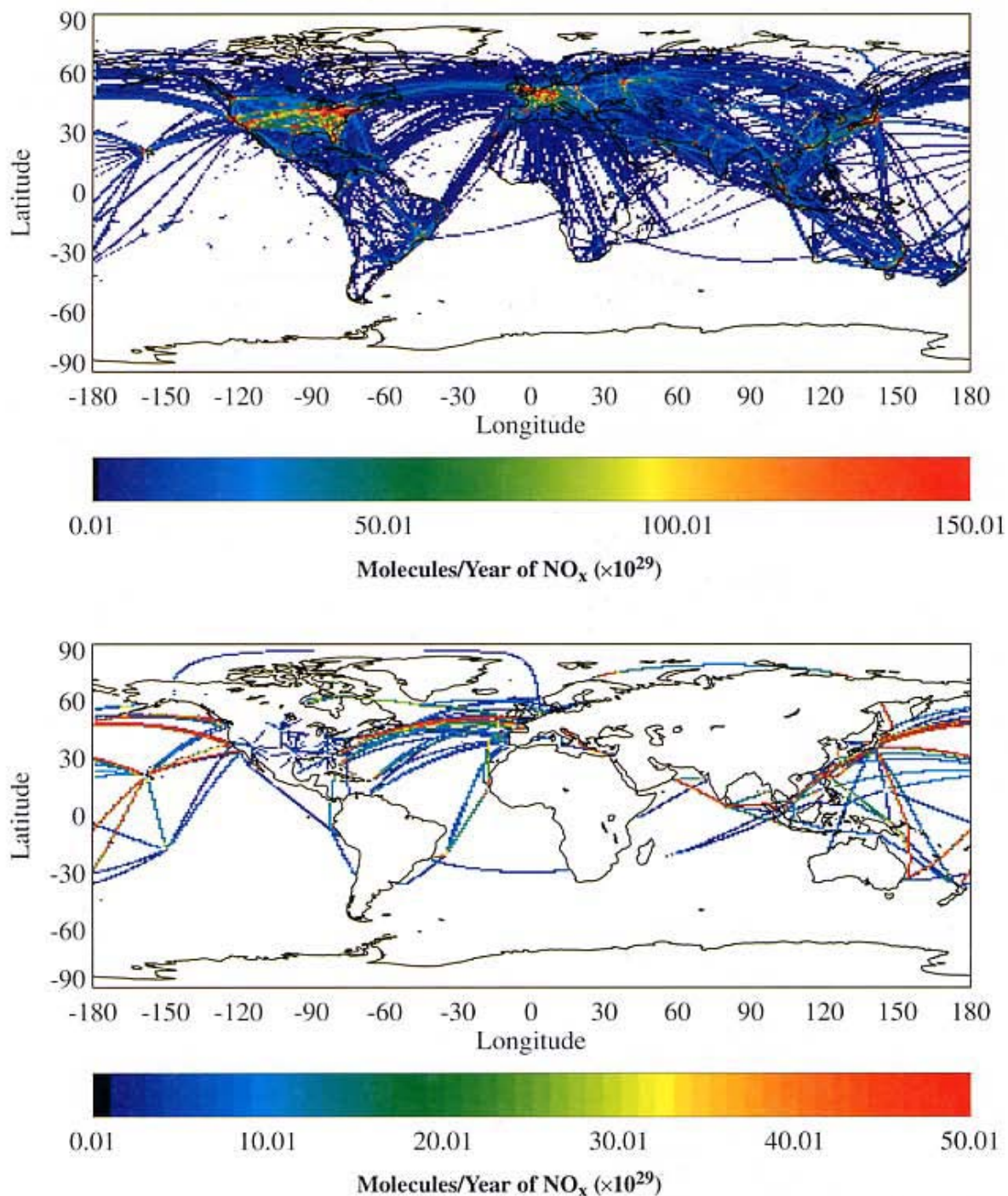


Figure 11-2. Annual NO_x emissions for proposed 2015 subsonic and Mach 2.4 ($\text{EI}(\text{NO}_x)=15$) HSCT fleets as function of latitude and longitude. Top panel shows emissions below 13 km (primarily subsonic traffic) while bottom panel shows emissions above 13 km (primarily HSCT traffic). (Albritton *et al.*, 1993)

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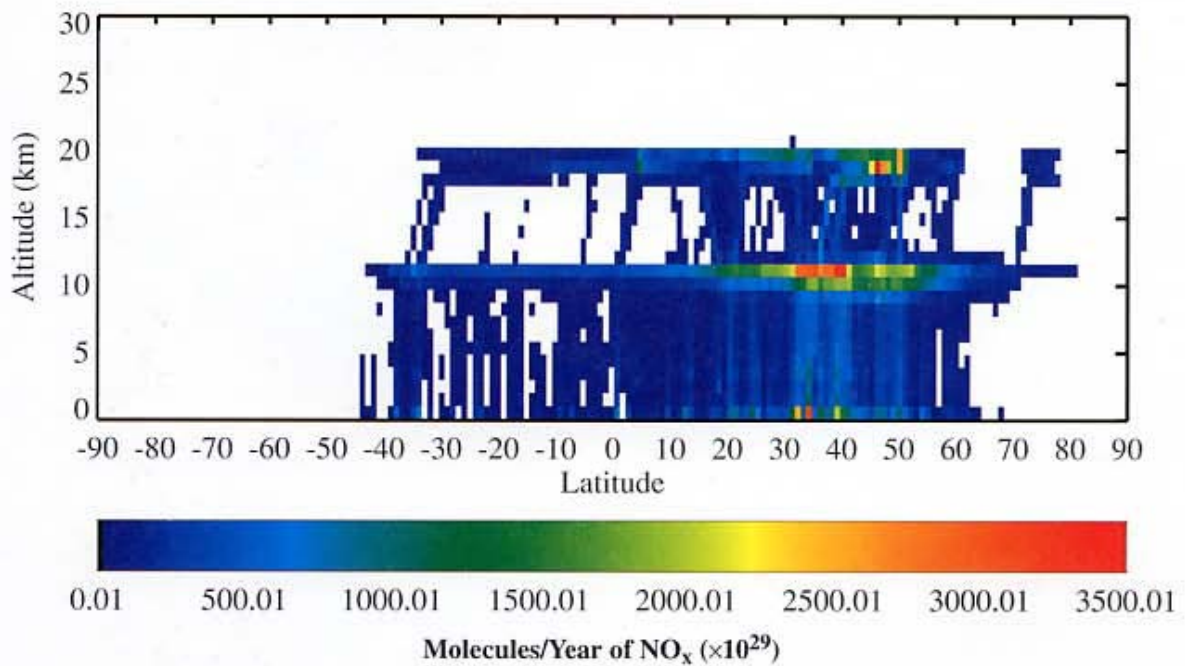
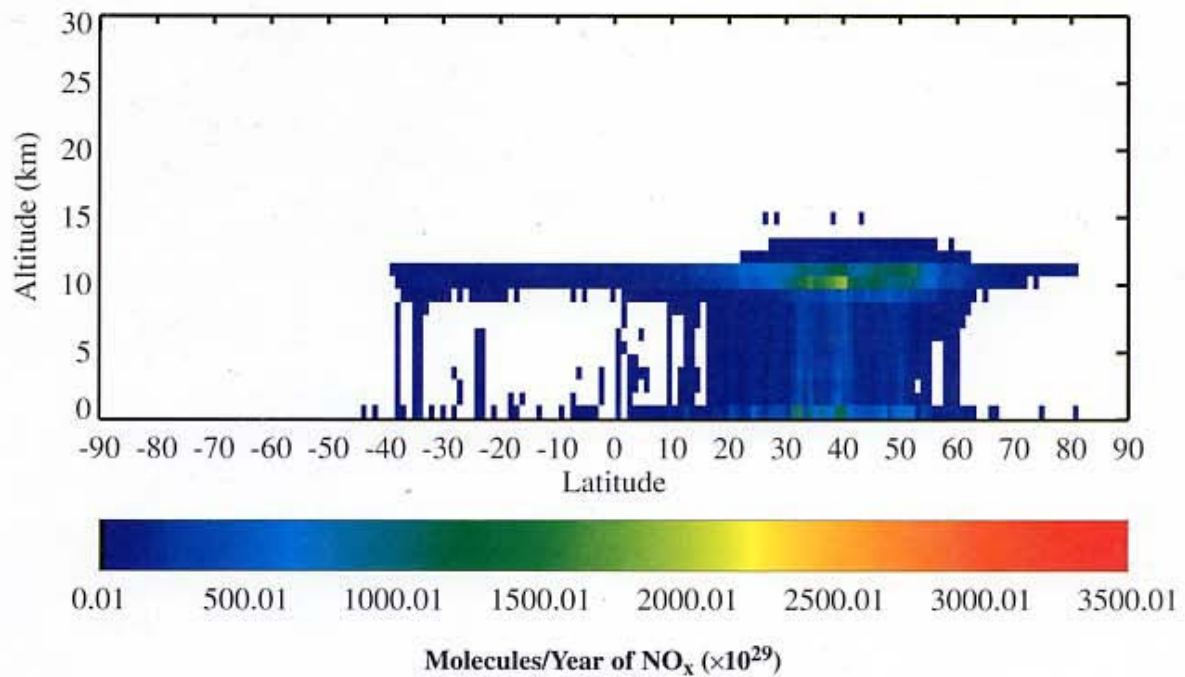


Figure 11-3. Annual NO_x emissions as a function of altitude and latitude for 1990 subsonic fleet (Scenario A, top panel) and for proposed 2015 subsonic and Mach 2.4 (EI(NO_x)=15) HSCT fleets (bottom panel). (Albritton *et al.*, 1993)

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(ECMWF) data, it is estimated that 44% of the NO_x emissions are injected in the lower stratosphere and 56% are injected in the upper troposphere.

11.3 PLUME PROCESSES

Plume processing involves the dispersion and conversion of aircraft exhausts on their way from the scales of the jet engines to the grid scales of global models. The details of plume mixing and processing can be important for conversion processes that depend nonlinearly on the concentration levels, such as the formation of contrails, the formation of soot, sulfur and nitric acid particles, and nonlinear photochemistry. Also, the vertical motion of the plumes relative to ambient air and sedimentation of particles may change the effective distribution of emitted species at large scales. Contrails may impact the mixing, sedimentation, heterogeneous chemistry, and the formation of cirrus clouds, with climatic consequences.

11.3.1 Mixing

The aircraft wake can be conveniently subdivided into three regimes (Hoshizaki *et al.*, 1975): the jet, the vortex, and the dispersion regimes. The vortex regime persists until the vortices become unstable and break up into a less ordered configuration. Thereafter, the dispersion regime follows, in which further mixing is influenced by atmospheric shear motions and turbulence depending on shear, stratification, and other parameters (Schumann and Gerz, 1993). With respect to mixing models, the jet and vortex regime, including the very early dispersion regime, can be computed with models as described by Miake-Lye *et al.* (1993). The engine plumes grow by turbulent mixing to fill the vortex pair cell. Due to rotation, centripetal acceleration causes inward motions of the relatively warm jet plumes so that the exhaust gases get trapped near the narrow well-mixed core of the vortices. The radial pressure gradient also causes adiabatic cooling and hence increases the formation of contrails. These centripetal forces are much larger for supersonic aircraft than for subsonic aircraft. It should be noted, however, that these model results remain largely untested, observationally.

Details of the plume fluid dynamics depend critically on the aircraft scales. For a Boeing-747, one may estimate that the jet regime lasts for about 10 s and the following vortex regime for about 1 to 3 minutes. The

cross-section of the trailing vortex pair represents an upper bound for the mixed area of the plumes. However, measurements of water vapor concentration and temperature in the jet and vortex regime (>2 km behind a DC-9 at cruising altitude) exhibit a spiky concentration field within the double vortex system, indicating that the individual jet plumes may not yet be homogeneously mixed over the vortex cross-section at such distances (Baumann *et al.*, 1993).

The lift of the aircraft induces downward motion of the double vortex structure at about $2.4 \pm 0.2 \text{ m s}^{-1}$ for a Boeing-747, which decreases when the vortices mix with the environment at altitudes that may be typically 100 m lower than flight level. During this descent, parts of the exhaust gases are found to escape the vortex cores.

In the supersonic case, the vortex pair has more vertical momentum (descent velocity of about 5 m/s), and its vertical motion will continue (possibly in the form of vortex rings) well after the vortex system has broken up. This will lead to exhaust species deposition a few hundred meters below flight altitude (Miake-Lye *et al.*, 1993). Radiation cooling of the exhaust gases may contribute to additional sinking (Rodriguez *et al.*, 1994), in particular when contrails are forming.

Very little is known about the rate of mixing in the dispersion range, and it is this rate of mixing that plays a large role in determining the time evolution of the gas composition of the plume (Karol *et al.*, 1994). In fact, it is yet unknown at what time scales the emissions become indistinguishable from the ambient atmosphere. Table 11-2 shows estimates of the concentration increases due to aircraft emissions in a young exhaust plume (vortex regime) and at the scales of the North Atlantic flight corridor (Schumann, 1994). These are the scales in between which global models will be able to resolve the concentration fields. The background concentration estimates are taken from Penner *et al.* (1991) for NO_x , Möhler and Arnold (1992) for SO_2 , and Pueschel *et al.* (1992) for soot. With respect to background, the concentration increases in young plumes are of importance for all aircraft emissions included in Table 11-2. A strong corridor effect is expected for NO_x and, at least in the lower stratosphere, also for SO_x and soot particles.

11.3.2 Homogeneous Processes

Several models have been developed to describe the finite-rate chemical kinetics in the exhaust plumes

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Table 11-2. Mean concentration increases in vortex regime (5000 m² cross-section) of a B-747 plume, and mean concentration increase in the North Atlantic flight corridor due to traffic exhaust emissions from 500 aircraft. (Table adopted from Schumann, 1994.)

| Species | EI (g/kg) | Background concentration at 8 km | Mean concentration increase in vortex regime | Mean concentration increase in North Atlantic flight corridor |
|------------------------------------|-----------|----------------------------------|--|---|
| CO ₂ | 3150 | 358 ppmv | 14 ppmv | 0.02 ppmv |
| H ₂ O | 1260 | 20-400 ppmv | 14 ppmv | 0.02 ppmv |
| NO _x (NO ₂) | 18 | 0.01-0.05 ppbv | 78 ppbv | 0.1 ppbv |
| SO ₂ | 1 | 50-300 pptv | 3100 pptv | 4 pptv |
| soot | 0.1 | 3 ng/m ³ | 240 ng/m ³ | 0.3 ng/m ³ |

(Danilin *et al.*, 1992; Miake-Lye *et al.*, 1993; Pleijel *et al.*, 1993; Weibrink and Zellner, 1993). Most models follow a well-mixed air parcel as a function of plume age or distance behind the aircraft. The models are initialized either with an estimate of emissions from the jet exit or a separate model describing the kinetics after the combustion chamber within the engine. Considerable deviations from local equilibrium are predicted at the jet exit, in particular for CO, NO, NO₂, HNO₃, OH, O, and H. In the models, the air parcel grows in size as a prescribed function of mixing with the environment, and the concentrations in the plume change according to mixing with the ambient air and due to internal reactions in the homogeneous mixture. The models differ in the treatment of mixing, in the reaction set used to simulate the exhaust plume finite-rate chemical kinetics, photolysis rates, treatment of heterogeneous processes, and in the prescription of the effective plume cross-section as a function of time or distance. Since most of the NO_x emissions are in the form of NO, a rapid but local destruction of ozone is to be expected.

Besides some incidental measurements in flight corridors or contrails (Hofmann and Rosen 1978; Douglass *et al.*, 1991), very few data exist at this time on the gaseous emissions in aircraft plumes in the atmosphere. Measurements of the gases HNO₂, HNO₃, NO, NO₂, and SO₂ were recently made (Arnold *et al.*, 1992, 1994a) in the young plume of an airliner at cruising altitude (see Figure 11-4). The data imply that not more than about

1% of the emitted odd-nitrogen underwent chemical conversion to longer living HNO₃. Hence, most of the emitted odd nitrogen initially remains in a reactive form, which can catalytically influence ozone.

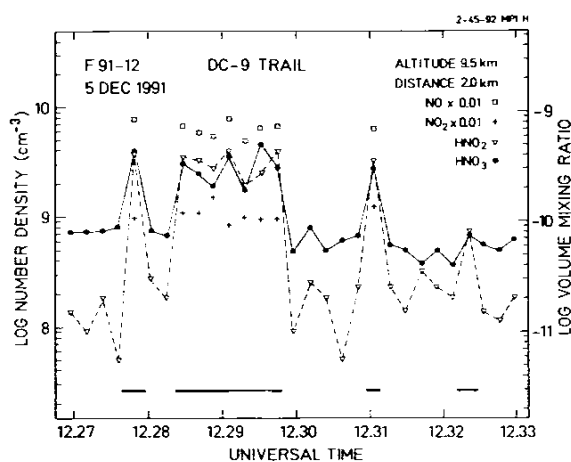


Figure 11-4. Time plot of nitrous acid (HNO₂) and nitric acid abundance measured during chase of a DC-9 airliner at 9.5 km altitude and a distance of 2 km. Periods when the research aircraft was inside the exhaust-trail of the DC-9 are marked by bars. For these periods NO and NO₂ abundance are also given. (Arnold *et al.*, 1992, 1994b; recalibration changed conversion factors shown in figure to: NO × 0.006 and NO₂ × 0.003.)

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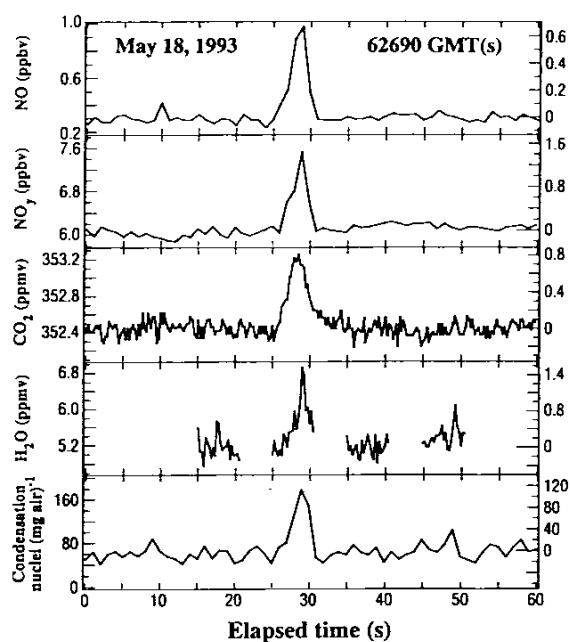


Figure 11-5. Time series for NO, NO_y, CO₂, H₂O, and CN during the plume encounters on May 18, 1993. The approximate Greenwich Mean Time (GMT) is noted in the top panel. The scale on the left side indicates the absolute value of each species. The zero in the right scale is set to the approximate background values of each species. At the ER-2 airspeed of 200 m s⁻¹, the panel width of 60 seconds corresponds to 12 km. (Based on Fahey *et al.*, 1994.)

In situ measurements of NO_y, NO, CO₂, H₂O, condensation nuclei, and meteorological parameters (Figure 11-5) have been used to observe the engine exhaust plume of the NASA ER-2 aircraft approximately 10 minutes after emission operating in the lower stratosphere (Fahey *et al.*, 1994). The obtained EI(NO_x) of 4 is in good agreement with values scaled from limited ground-based tests of the ER-2 engine. Non-NO_x nitrogen species comprise less than about 20% of emitted reactive nitrogen, consistent with model evaluations.

11.3.3 Heterogeneous Processes

New particles form in young exhaust plumes of jet aircraft. This is documented by *in situ* condensation nucleus (CN) measurements made (Hofmann and Rosen,

1978; Pitchford *et al.*, 1991; Hagen *et al.*, 1992; Whitefield *et al.*, 1993) in plumes under flight conditions.

The molecular physics details of nucleation are not well known and the theory of bimolecular nucleation is only in a rudimentary state. For a jet engine exhaust scenario, nucleation takes place in a non-equilibrium mechanism, which further complicates a theoretical description. It seems, however, that jet aircraft may form long-lived contrails composed of H₂SO₄·H₂O aerosols and soot particles covered with H₂SO₄·H₂O. Under conditions of low ambient temperatures around 10 km altitude, particularly in winter at high latitudes, contrails composed of HNO₃·H₂O aerosols may also form (Arnold *et al.*, 1992). Even if HNO₃·H₂O nucleation does not occur, some HNO₃ may become incorporated into condensed-phase H₂SO₄·H₂O by dissolution at low temperatures.

There are several potential effects of newly formed CN and activated soot. Such CN may trigger water contrail formation, induce heterogeneous chemical reactions, and serve as cloud condensation nuclei (CCN). Thereby, jet aircraft-produced CN may have an impact on trace gas cycles and climate. However, at present this is highly speculative.

Numerical calculations with chemical plume models show that the impact of aircraft emissions on the atmosphere in the wake regime critically depends on heterogeneous processes where considerable uncertainties still exist (Danilin *et al.*, 1992, 1994). Danilin *et al.* (1992) have considered the heterogeneous reaction N₂O₅ + H₂O → 2HNO₃ on ambient aerosol particles only. They have found that this reaction does not play an important role at time scales of up to one hour in the wake, but may get important at larger time scales. Taking contrail ice (or/and nitric acid trihydrate [NAT]) particle formation into account, Danilin *et al.* (1994) estimate that heterogeneous processes are more important at lower temperatures, but their impact on heterogeneous conversion is small during the first day after emission. In contrast, Karol *et al.* (1994) found noticeable "heterogeneous impact" on the chemistry in the plume taking into account the growth of ice particles.

Around 10 km altitude, there seems to exist a strong CN source, which is not due to aircraft but to H₂SO₄ resulting from sulfur sources at the Earth's surface (Arnold *et al.*, 1994a). Hence, the relative contribution of aircraft to CN production around 10 km

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Table 11-3. Estimates of stratospheric perturbations due to aircraft effluents of a fleet of approximately 500 Mach 2.4 HSCTs (NO_x EI=15) relative to background concentrations. (Perturbations are estimated for a broad corridor at northern midlatitudes.) (Expanded from Stolarski and Wesoky, 1993b.)

| Species | Perturbation | Background |
|--------------------------------|---------------|------------------------------|
| NO _x | 3-5 ppbv | 2-16 ppbv |
| H ₂ O | 0.2-0.8 ppmv | 2-6 ppmv |
| SO _x | 10-20 pptv | 50-100 pptv |
| H ₂ SO ₄ | 350-700 pptm | 350-700 pptm |
| Soot | ~7 pptm | ~7 pptm |
| Hydrocarbons | 2 ppbv (NMHC) | 1600 ppbv (CH ₄) |
| CO | ~2ppbv | 10-50 ppbv |
| CO ₂ | ~1 ppbv | 350 ppmv |

altitude needs to be determined. It is uncertain whether CN production around 10 km actually has a significant impact on trace gas cycles and CCN.

11.3.4 Contrails

Miake-Lye *et al.* (1993) have applied the analysis of Appleman (1953) to the standard atmosphere as a function of altitude and latitude. Their result shows that much of the current high-flying air traffic takes place at altitudes where the formation of contrails is very likely, in particular in the northern winter hemisphere. A small reduction of global mean temperature near and above the tropopause, by say 2 K, would strongly increase the region in which contrails have to be expected. Also, a slight change in the threshold temperature below which contrails form has a strong effect on the area of coverage with contrails.

Except for *in situ* measurements by Knollenberg (1972), little is known about the spatial structure and microphysical parameters of contrails. Recent measurements (Gayet *et al.*, 1993) show that contrails contain more and smaller ice particles than natural cirrus, leading to about double the optical thickness in spite of their smaller ice content. Contrail observations from satellite data, Lidar measurements, and climatological observations of cloud cover changes have been described by Schumann and Wendling (1990). Large (1 to 10 km wide and more than 100 km long) contrails are observed regionally on about a quarter of all days within one year, but the average contrail coverage is only about 0.4% in mid-Europe. Lidar observations show that particles from

contrails sediment quickly at approximately 10 km altitude (Schumann, 1994).

11.4 NO_x/H₂O/SULFUR IMPACTS ON ATMOSPHERIC CHEMISTRY

11.4.1 Supersonic Aircraft

The impacts of HSCT emissions on chemistry are discussed in detail in Stolarski and Wesoky (1993b). Here we give a short summary. Effects of emissions from HSCTs (see Table 11-3) on ozone are generally predicted to be manifested through gas phase catalytic cycles involving NO_x, HO_x, ClO_x, and BrO_x. The amounts of these radicals are changed by two pathways. First, they are changed by chemistry, either addition of or repartitioning within nitrogen, hydrogen, and halogen chemical families. Predicted changes in ozone from this pathway are initiated primarily by NO_x chemistry. Second, they are changed when HSCT emissions affect the properties of the aerosols and the probability of polar stratospheric cloud (PSC) formation. Changes in ozone from this pathway are determined primarily by ClO_x and BrO_x chemistry, with a contribution from HO_x chemistry (see Chapter 6 for more detail).

Heterogeneous chemistry on sulfate aerosols also has a large impact on the potential ozone loss. Most important is the hydrolysis of N₂O₅: N₂O₅ + H₂O → 2 HNO₃. Several observations are consistent with this reaction occurring in the lower stratosphere (*e.g.*, Fahey *et al.*, 1993; Solomon and Keys, 1992). Its most direct

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effect is to reduce the amount of NO_x . Indirectly, it increases the amounts of ClO and HO_2 by shifting the balance of ClO and ClONO_2 more toward ClO during the day and by reducing the loss of HO_x into HNO_3 . As a result, the HO_x catalytic cycle is the largest chemical loss of ozone in the lower stratosphere, with NO_x second, and both the ClO_x and BrO_x catalytic cycles have increased importance compared to gas phase conditions.

The addition of the emissions from HSCTs will affect the partitioning of radicals in the NO_y , HO_y , and ClO_y chemical families, and thus will affect ozone. The NO_x emitted from the HSCTs will be chemically converted to other forms, so that the NO_x/NO_y ratio of these emissions will be almost the same as for the background atmosphere. As a result, the NO_x emissions will tend to decrease ozone, but less than would occur in the absence of sulfate aerosols.

The increase in H_2O will lead to an increase in OH , because the reaction between $\text{O}(\text{ID})$ that comes from ozone photolysis and H_2O is the major source of OH ; however, increases in NO_y will act to reduce HO_x through the reactions of OH with HNO_3 and HNO_4 . On the other hand, HNO_3 , formed in the reaction of OH with NO_2 , can be photolyzed in some seasons and latitudes to regenerate OH . When all of these effects are considered, the amount of HO_x is calculated to decrease— HO_2 by up to 30% and OH by up to 10%. Thus, the catalytic destruction of ozone by HO_x , the largest of the catalytic cycles, is decreased.

Finally, ClO_x concentrations decrease with the addition of HSCT emissions for two reasons. First and most important, with the addition of more NO_2 , the daytime balance between ClO and ClONO_2 is shifted more toward ClONO_2 . Second, with OH reduced, the conversion of HCl to Cl by reaction with OH is reduced, so that more chlorine stays in the form of HCl . Thus, the catalytic destruction of ozone by ClO_x is decreased.

The addition of HSCT emissions results in increases in the catalytic destruction of ozone by the NO_x cycle that are compensated by decreases in the catalytic destruction by ClO_x and HO_x . Because the magnitudes of the changes in catalytic destruction of ozone are similar for the NO_x , HO_x , and ClO_x cycles, compensation results in a small increase or decrease in ozone. Model calculations indicate a small decrease. The decreases in the catalytic destruction of O_3 by ClO_x and HO_x involve the effects of increased water vapor and HNO_3 on the

rates of heterogeneous reactions on sulfate and the probability of PSC formation.

The addition of sulfur to the stratosphere from HSCTs will increase the surface area of the sulfate aerosol layer. This change in aerosol surface area is expected to be small compared to changes from volcanic eruptions, with a possible exception being the immediate vicinity of the aircraft wake. Model calculations by Bekki and Pyle (1993) predict regional increases of the mass of lower stratospheric $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ aerosols, due to air traffic, by up to about 100%. The importance of sulfur emissions from HSCTs in the presence of this large and variable background needs to be assessed.

11.4.2 Subsonic Aircraft

The emissions from subsonic aircraft take place both in the lower stratosphere and troposphere. The primary chemical effects of aircraft in the troposphere seem to be related to their NO_x emissions. The concentration of ozone in the upper troposphere depends on transport of ozone mainly from the stratosphere and on upper troposphere ozone production or destruction. The impact of subsonic aircraft occurs through the influence of NO_x on the tropospheric HO_x cycle (see Chapter 5 for a fuller discussion of tropospheric ozone chemistry).

The HO_x cycle is initialized by the photolysis of ozone itself, which results in the production of OH radicals and destruction of ozone. OH radicals have two possible reaction pathways: reaction with CO , CH_4 , and non-methane hydrocarbons (NMHC) resulting in HO_2 and RO_2 radicals; or reaction with NO_2 , removing OH and NO_x from the cycle. The HO_2 radicals that are produced also have two possible pathways: reaction with ozone or reaction with NO . The first one removes ozone from the cycle; the second one (also valid for RO_2 radicals) produces ozone and regains NO . Additionally, both pathways regain OH radicals.

As a consequence, ozone is destroyed photochemically in the absence of NO_x . Only in the presence of NO_x can ozone be produced. The net production/destruction depends on the combination of these processes. Their relative importance is controlled mainly by the NO_x concentration. In a regime of low NO_x , the ozone concentration will be reduced photochemically. At higher NO_x concentrations (on the order of 10 pptv NO_x) NO_x will lead to a net ozone production. In both regimes, additional NO_x will result in higher ozone

concentrations. Only when the concentration of NO_x is so high (over a few hundred pptv NO_x) that the OH concentration starts to decline, will additional NO_x result in a lower ozone production.

The impact of NO_x emitted by aircraft depends, therefore, on the background NO_x concentration and on the increase in NO_x concentration. Measurements show that background NO_x concentrations (including NO_x emitted from subsonic aircraft) are in the range of 10-200 pptv NO_x . Therefore, airplane emissions take place in the regime of increasing ozone production most of the time, where increasing NO_x results in increased local ozone concentrations.

In this regime, the concentration of OH radicals is enhanced also by additional NO_x . First, enhanced ozone means higher production of OH by photolysis of ozone. Second, the partitioning in the HO_x family is shifted towards OH by the reaction of HO_2 with NO. The loss process of OH by reaction with NO_2 is not yet important. This enhancement of the OH concentration reduces the tropospheric lifetime of many trace species like CH_4 , NO_x , etc.

The emission of sulfur from aviation is much smaller than from surface emissions and negligible in terms of the resultant acid rain, but may be important if emitted at high altitudes. Hofmann (1991) reported observations that show an increase of non-volcanic stratospheric sulfate aerosol of about 5% per year. He suggests that if about 1/6 of the Northern Hemisphere air traffic takes place directly in the stratosphere and if a small fraction of other emissions above 9 km would enter the stratosphere through dynamical processes, then the jet fleet appears to represent a large enough source to explain the observed increase. On the other hand, Bekki and Pyle (1992) conclude from a model study that although aircraft may represent a substantial source of sulfate below 20 km, the rise in air traffic is insufficient to account for the observed 60% increase in large stratospheric aerosol particles over the 1979-1990 period. Sulfate particles generated from SO_x may also contribute to nucleation particles (Arnold *et al.*, 1994a). Whitefield *et al.* (1993) find a positive correlation between sulfur content and CCN efficiency of particles formed in jet engine combustion.

The possible enhancement of aerosol surface area may affect the nighttime chemistry of the nitrogen oxides. The heterogeneous reaction of N_2O_5 (and possibly

NO_3) on aerosol surfaces will reduce the concentration of photochemically active NO_x during the day, giving rise to lower ozone and OH concentrations in the upper troposphere (Dentener and Crutzen, 1993).

11.5 MODEL PREDICTIONS OF AIRCRAFT EFFECTS ON ATMOSPHERIC CHEMISTRY

The first investigations concerning the potential effects of supersonic aircraft on the ozone layer were conducted in the 1970s. Early assessments were obtained using one-dimensional (1-D) photochemical models; more recent assessments rely on 2-D models (*e.g.*, Stolarski and Wesoky, 1993b). In addition, the transport in 2-D models has been compared to 3-D model transport by examining the evolution of the distribution of passive tracers.

11.5.1 Supersonic Aircraft

Evaluations of the effects of the emissions of the HSCT on the lower stratosphere have used two-dimensional (2-D) models. These are zonally averaged (latitude-height) models and are discussed in detail in Chapter 6. For use in such 2-D models, both the source of exhaust and the emission transport (both horizontal and vertical) are zonally averaged. In fact, the source of emissions is not zonally symmetric, as HSCT flight is expected to be restricted to oceanic corridors. Furthermore, the transport processes through which trace species are removed from the stratosphere are not well represented by a zonally averaged model. Stratosphere-troposphere exchange processes (STE) occur preferentially near jet-systems, above frontal perturbations, and during strong convection in tropical regions. The two former processes may transport effluents released by HSCTs irreversibly to lower levels and lead to tropospheric sinks. Effluents may be rapidly advected also to lower latitudes by large-scale motions. Such processes are poorly represented in 2-D models. The horizontal scale for STE is small and can only be represented using 3-D models with high resolution. These small scales are not explicitly resolved in most global 3-D models. Thus, any use of a 3-D model to evaluate the use of a 2-D model for these assessments must include a critical evaluation of the 3-D model STE. 2-D models do have the practical advantage that it is possible to complete many

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assessment calculations, using a reasonably complete representation of stratospheric chemistry, and also by considering the sensitivity of the results to model parameters one can take some aspects of feedbacks among atmospheric processes into account.

Current 3-D models, though impractical for full chemical assessments, are practical for calculations that consider the transport of aircraft exhaust, which is treated as a passive tracer. Such calculations have been compared directly with 2-D models (Douglass *et al.*, 1993; Rasch *et al.*, 1993). Their results show that for seasonal simulations, provided that the residual circulation derived from the 3-D fields is the same as used in the 2-D calculation, the tracer is dispersed faster vertically and has similar horizontal spread for 3-D compared with 2-D calculations. Although the tracer is also transported upward more rapidly in 3-D than in 2-D (where vertical upward transport is minimal), the more rapid downward transport is the more pronounced effect. Accumulation of aircraft exhaust in flight corridors is found in regions of low wind speed, but only a small number of typical corridors (North Atlantic, North Pacific, and tropical) have been considered. The effect of such local accumulation would be largest if a threshold chemical process such as particle formation is triggered at high concentration of aircraft exhaust constituents. In 2-D models that use residual mean formulation, transport to the troposphere takes place principally through two mechanisms: advective transport by the residual mean circulation (mostly at middle to high latitudes) and diffusive transport across the tropopause (all latitudes). The latter is largest where the 2-D model's tropopause height is discontinuous (to represent the downward slope of the tropopause from the tropics to middle and polar latitudes) (Shia *et al.*, 1993). The difference in the character of STE in 2-D and 3-D models leads to different sensitivities to the latitude at which exhaust is injected in the models. For the 3-D model, the atmospheric lifetime of a tracer species is relatively insensitive to the latitude of injection. For the 2-D model, the tracer species lifetime is much longer for injection at lower latitudes than at higher latitudes, since transport to higher latitudes must take place before most of the pollutant is removed from the stratosphere.

Treatments of the transport and photochemistry used in 2-D models have been examined through a series of model intercomparisons and comparisons with obser-

vations (Jackman *et al.*, 1989b; Prather and Remsberg, 1993). Model results for a "best" simulation, as well as for various applications and constrained calculations, were compared with each other and with observations. There are significant differences in the models that lead to differences in the model assessments as discussed below. In addition, there are some features, such as the very low observed values of N_2O and CH_4 in the upper tropical stratosphere, and the NO_y/O_3 ratio at tropical latitudes, that are not well represented by all 2-D models.

There are also many areas of agreement between models and observations that suggest that an evaluation of the effects of the HSCT may be an appropriate use of these models. For example, the models' total ozone fields show general consistency when compared with observed fields such as Total Ozone Mapping Spectrometer (TOMS) data, the overall vertical and latitudinal distributions of such species as N_2O , CH_4 , and HNO_3 , and the ozone climatology that is based on Stratospheric Aerosol and Gas Experiment (SAGE) and Solar Backscatter Ultraviolet (SBUV) observations. If the SAGE results for O_3 loss over the past decade at altitudes just above the tropopause are correct (see Chapter 1), however, then the inability of present models to reproduce this O_3 decrease (see Chapter 6) casts doubt on their ability to correctly model aircraft effects in this important region.

At the beginning of the NASA HSRP/AESA program, the assessment models contained only gas phase photochemical reactions. The importance of the heterogeneous reaction (temperature independent) $N_2O_5 + H_2O \rightarrow 2 HNO_3$ on the surface of stratospheric aerosols was noted by Weisenstein *et al.* (1991) and Bekki *et al.* (1991) and has been further explored by Ramarason and Louisnard (1994). This process changes the balance between the reactive nitrogen species, NO and NO_2 (NO_x), and the reservoir species, HNO_3 . For gas phase evaluations, lower stratospheric ozone was most sensitive to the amount of NO_x from aircraft exhaust injected into the lower stratosphere. For evaluations including this heterogeneous process, the NO_x levels in both the base atmosphere and in the perturbed atmosphere are much lower than in the gas phase evaluations, and the calculated ozone change is greatly reduced (Ko and Douglass, 1993).

2-D models have also been used to examine other processes that are of potential significance. For example,

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Table 11-4. Calculated percent change in the averaged column content of ozone between 40°N and 50°N.

| Scenarios | AER | GSFC | LLNL | OSLO | CAMED | NCAR |
|--------------------------------------|-------|-------|-------|-------|-------|-------|
| I: Mach 1.6, NO _x EI=5* | -0.04 | -0.11 | -0.22 | +0.04 | +0.69 | -0.01 |
| II: Mach 1.6, NO _x EI=15* | -0.02 | -0.07 | -0.57 | +0.15 | +0.48 | -0.60 |
| III: Mach 2.4, NO _x EI=5* | -0.47 | -0.29 | -0.58 | -0.47 | +0.38 | -0.26 |
| IV: Mach 2.4, NO _x EI=15* | -1.2 | -0.86 | -2.1 | -1.3 | -0.45 | -1.8 |
| V: Mach 2.4, NO _x EI=15** | -2.0 | -1.3 | -2.7 | -0.42 | -1.1 | -2.3 |
| VI: Mach 2.4, NO _x EI=45* | -5.5 | -4.1 | -8.3 | -3.5 | -2.8 | -6.9 |

Table 11-5. Calculated percent change in the averaged column content of ozone in the Northern Hemisphere.

| Scenarios | AER | GSFC | LLNL | OSLO | CAMED | NCAR |
|--------------------------------------|-------|-------|-------|-------|-------|-------|
| I: Mach 1.6, NO _x EI=5* | -0.04 | -0.12 | -0.18 | +0.02 | +0.63 | -0.04 |
| II: Mach 1.6, NO _x EI=15* | -0.02 | -0.14 | -0.48 | +0.10 | +0.63 | -0.54 |
| III: Mach 2.4, NO _x EI=5* | -0.42 | -0.27 | -0.50 | -0.39 | +0.25 | -0.25 |
| IV: Mach 2.4, NO _x EI=15* | -1.0 | -0.80 | -1.8 | -1.0 | -0.26 | -1.5 |
| V: Mach 2.4, NO _x EI=15** | -1.7 | -1.2 | -2.3 | -0.43 | -0.80 | -1.9 |
| VI: Mach 2.4, NO _x EI=45* | -4.6 | -3.6 | -7.0 | -3.1 | -2.1 | -5.1 |

* Relative to a background atmosphere with chlorine loading of 3.7 ppbv, corresponding to the year 2015

** Relative to a background atmosphere with chlorine loading of 2.0 ppbv, corresponding to the year 2060

if HSCT planes are flown, the lower stratospheric levels of total odd nitrogen and water vapor are expected to rise. In addition to a general increase over background levels throughout the lower stratosphere, there is a possibility for large enhancements in areas of high traffic (air "corridors"). Peter *et al.* (1991) and Considine *et al.* (1993) have considered the possibility that the increases in H₂O and in HNO₃ (a consequence of the heterogeneous conversion of NO_x) will lead to an increase in the amount of nitric acid trihydrate (NAT) cloud formation. They indeed find this to be so.

The evaluation of the effects of a future fleet of supersonic aircraft on stratospheric ozone was made by Johnston *et al.* (1989) and by Ramarosan (1993) using gas phase models. The ozone loss for an injection at a fixed level was found to increase nearly linearly as the amount of NO_x injected was increased. The ozone loss was found to be larger for injection at higher levels because the ozone response time decreases with altitude, and because the pollutant has a longer stratospheric lifetime when injected farther from the model tropopause.

Jackman *et al.* (1989a) used a 2-D model to test the dependence of the supersonic aircraft assessments on model dynamical inputs. As anticipated, the calculated change in ozone is larger (smaller) for a slower (faster) residual circulation because the circulation controls the magnitude of the steady-state stratospheric NO_x pertur-

bation. This paper also showed that the annual cycle of the zonally averaged total ozone is sensitive to the annual cycle in the residual circulation. A similar sensitivity to the residual circulation has been demonstrated for a 3-D calculation using winds from a data assimilation procedure for transport (Weaver *et al.*, 1993).

The supersonic aircraft assessment scenarios discussed here are for Mach numbers 1.6 and 2.4, which correspond to the two aircraft cruise altitudes 16 km and 20 km, respectively, and for three values for EI(NO_x) (see Stolarski and Wesoky [1993b] for specific details). The emission indices are given in Table 11-1. The calculated total ozone changes are given for each participating model in Table 11-4 for the calculated annually averaged column ozone change in the latitude band where the aircraft emissions are largest (40°-50°N), and in Table 11-5 for the Northern Hemisphere average. The model calculations use an aerosol background similar to that observed in 1979 (*e.g.*, before the Mt. Pinatubo eruption). Some similarities and differences are seen among the model results. For all of the models, the ozone change for Mach 2.4 is more negative than that for Mach 1.6. The ozone change at Mach 2.4 is more negative as the EI is increased, but the change is more rapid than a linear change. The complexity of the assessment is captured by the change in ozone calculated at Mach 1.6 for the two different EIs in Table 11-4. For all models,

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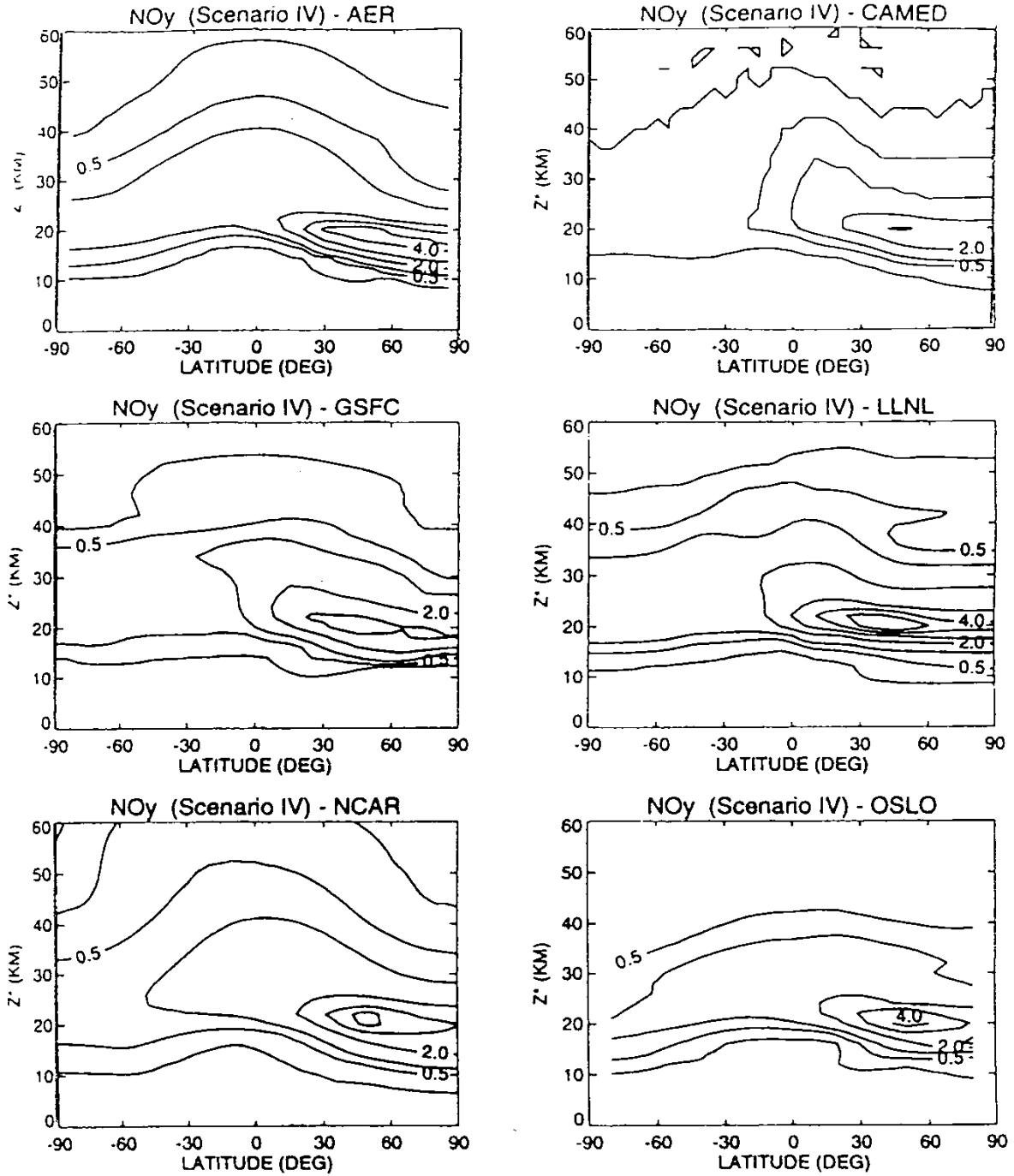


Figure 11-6. Calculated changes in the local concentration of NO_y (ppbv) in June for Mach 2.4 (EI(NO_x)=15) case. The contour intervals are 1 ppbv, 2 ppbv, 3 ppbv, 4 ppbv, and 5 ppbv (Stolarski and Wesoky, 1993b).

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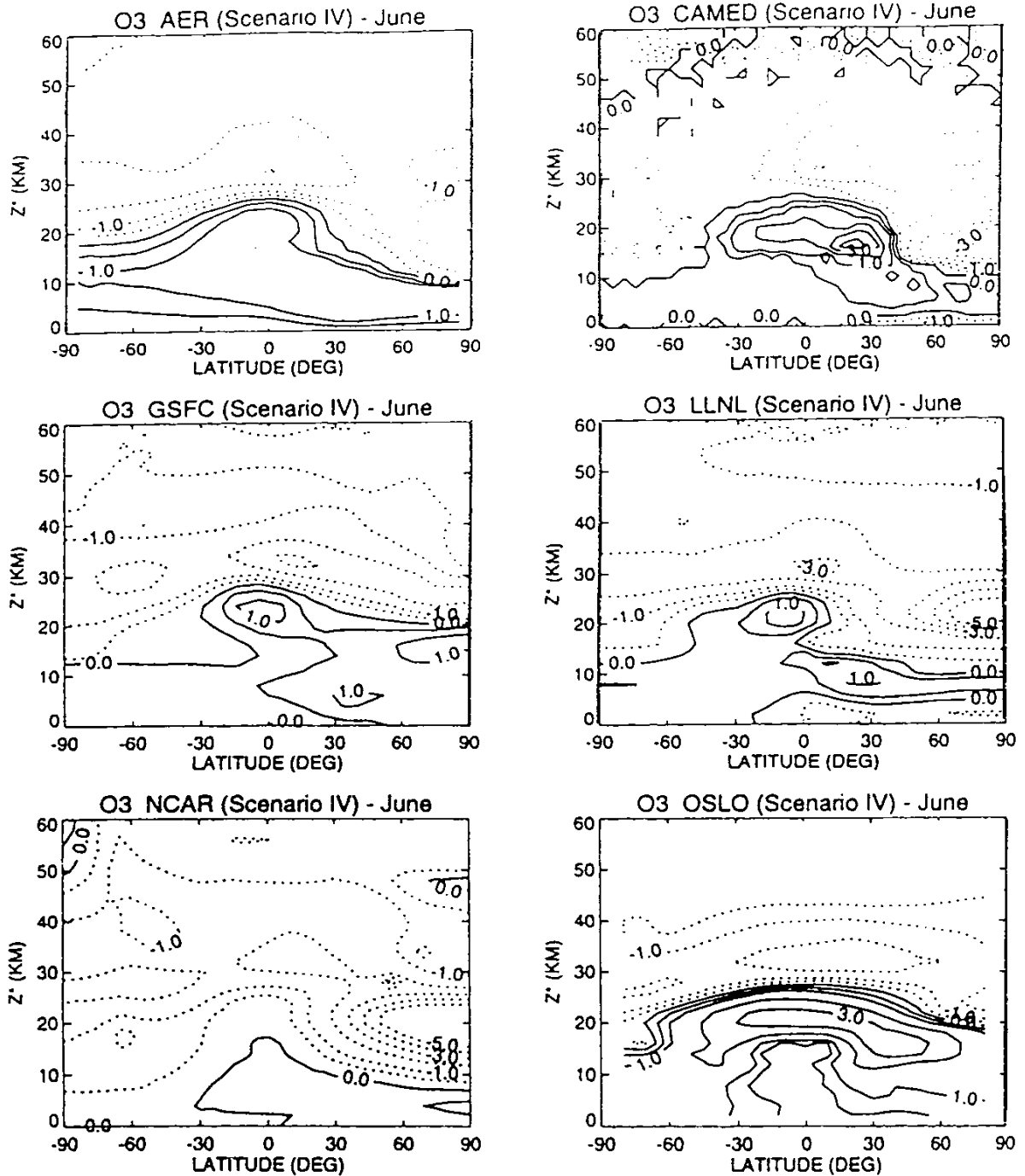


Figure 11-7. Model-calculated percent change in local ozone for June for Mach 2.4 ($EI(NO_x)=15$) fleet in the 2015 atmosphere. The contour intervals are -4%, -3%, -2%, -1%, -0.5%, 0%, 0.5%, 1%, 2%, 3%, 4% (Albritton *et al.*, 1993).

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and for both cases at ClO_x mixing ratios of 3.7 ppbv, the changes are less than 1%. For three of the models (Atmospheric and Environmental Research, Inc., AER; Goddard Space Flight Center, GSFC; and the University of Oslo, OSLO), the ozone change is less negative (more positive) for $\text{EI} = 15$ than for $\text{EI} = 5$. For the other three models (Lawrence Livermore National Laboratory, LLNL; the University of Cambridge and the University of Edingburgh, CAMED; and the National Center for Atmospheric Research, NCAR), the ozone change is more negative (less positive) for the larger emission index.

The assessment initiated by the "Comite Avion-Ozone" shows similar results. A 2-D model including heterogeneous reactions on aerosol and PSC surfaces and a similar emission scenario to that for the HSRP assessments shows a global mean decrease of total ozone of 0.3% (Ramaroson and Louisnard, 1994). The results depend upon the prescribed background atmosphere (e.g., aerosol loading) used (see also: Tie *et al.*, 1994; Considine *et al.*, 1994).

The change in NO_y is given in Figure 11-6 for each of the models for a scenario in which the HSCT fleet is assumed to fly at Mach 2.4 with an $\text{EI}(\text{NO}_x) = 15$ and a background chlorine mixing ratio of 3.7 ppbv. This NO_y change indicates the sensitivity to the different transport. LLNL has the largest change in NO_y , and also the largest global ozone changes in Tables 11-4 and 11-5. However, the calculated global changes are clearly not ordered by the magnitude of the NO_y change. The latitude height change in ozone for each of the models is given in Figure 11-7. There are remarkably large differences in the local ozone changes, particularly in the upper troposphere/lower stratosphere region where the aircraft emissions produce an increase in the ozone production as well as an increase in the ozone loss. Although changes in NO_x have the largest impact on O_3 , the effects from H_2O emissions contribute to the calculated O_3 changes (about 20%).

The assessment models' representation of upper tropospheric chemistry was not considered as a part of the Models and Measurements Workshop (Prather and Remsberg, 1993). Further attention must be paid to the upper tropospheric chemistry to understand the spread in the results for these assessments. This subject is discussed in the following section on the evaluation of the impact of the subsonic fleet.

11.5.2 Subsonic Aircraft

The Chapter 7 discussions indicate that tropospheric photochemical-dynamic modeling is much less developed than is this type of stratospheric modeling; however, several types of models have been used to assess the impact of subsonic aircraft emissions. These include global photochemistry and transport models in latitude-height dimensions ignoring the longitudinal variation of emissions. This is an important drawback for species with short lifetimes. Another type of model used is the longitude-height model that addresses a restricted range of latitudes. They neglect the effect of latitudinal transport. Three-dimensional global dynamical models are being developed to study the impact of aircraft emissions, but the results from these models are as yet restricted to NO_x and NO_y species. The published results from two-dimensional models have used a range of estimates to represent present and future aircraft emissions, and consequently, the results are not easily comparable. There have been no organized efforts to intercompare models for subsonic aircraft as there have been for the supersonic aircraft problem.

The sensitivity of modeled ozone concentrations to changes in aircraft NO_x emissions has been found to be much higher than for surface emissions, with around twenty times more ozone being created per unit NO_x emission for aircraft compared to surface sources (Johnson *et al.*, 1992). Several authors have investigated the role of hydrocarbon and carbon monoxide emissions from aircraft on ozone concentrations, but have found small effects (Beck *et al.*, 1992; Johnson and Henshaw, 1991; Wuebbles and Kinnison, 1990). The increase in net ozone production with increasing NO_x is steeper at lower concentrations of NO_x (Liu *et al.*, 1987), and therefore larger ozone sensitivities are expected for emissions to the Southern Hemisphere, where NO_x concentrations are lower (Johnson and Henshaw, 1991). Beck *et al.* (1992) note the influence of lightning production of NO_x in controlling the sensitivity of ozone to aircraft NO_x emissions. These studies indicate the importance of predicting a realistic background NO_x concentration, and underline the importance of measurements in model testing.

Several recent publications (Johnson and Henshaw, 1991; Wuebbles and Kinnison, 1990; Fuglested *et al.*, 1993; Beck *et al.*, 1992; Rohrer *et al.*, 1993) esti-

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mate the percentage increases in ozone concentrations due to the impact of aircraft emissions. The results show maximum increases at around 10 km of between 12% and 4% between 30° and 50°N.

NO_x concentrations in the upper troposphere are controlled by the transport of NO_x downwards from the stratosphere, by aircraft and lightning emissions, and by the convection of NO_x from surface sources (Ehhalt *et al.*, 1992). The available measurements of NO_x in the free troposphere are discussed in Chapter 5. There are a number of observations where the vertical NO profile is strongly and unequivocally influenced by one or the other of these sources, *e.g.*, lightning (Chameides *et al.*, 1987; Murphy *et al.*, 1993), aircraft emissions (Arnold *et al.*, 1992), fast vertical transport (Ehhalt *et al.*, 1993), which makes it clear that all these sources can and do make a contribution to the NO_x in the upper troposphere. An example is given in Figure 11-8, which presents the daytime NO distribution across the North Atlantic during the period June 4-6, 1984, of the Stratospheric Ozone (STRATOZ III) campaign (Ehhalt *et al.*, 1993). Large longitudinal gradients of NO mixing ratio up to a factor of 5 were observed at all altitudes in the free troposphere in which the effects of an outflow of polluted air from the European continent are seen. This tongue of high NO over the Eastern Atlantic was accompanied by elevated CO and CH₄ mixing ratios and therefore was probably due to surface sources. Figure 11-8 also illustrates the variance superimposed by longitudinal gradients on average meridional cross sections. However, at present there are not enough data to derive the respective global contributions from atmospheric measurements alone. Independent estimates of the various source strengths are needed. Our lack of knowledge about the NO_x budget in the troposphere, especially in the upper troposphere, makes model predictions for this region questionable. Thus, at present, we can have little confidence in our ability to correctly model subsonic aircraft effects on the atmosphere.

Figure 11-9 shows published comparisons of available NO measurements (Wahner *et al.*, 1994) with predictions from two-dimensional models (Berntsen and Isaksen, 1992). Using a quasi-two dimensional longitude-height model and considering estimates of all important tropospheric sources of NO_x (input from the stratosphere, lightning, fossil fuel combustion, soil emissions and aircraft) for the latitude band of 40°-50°N (see

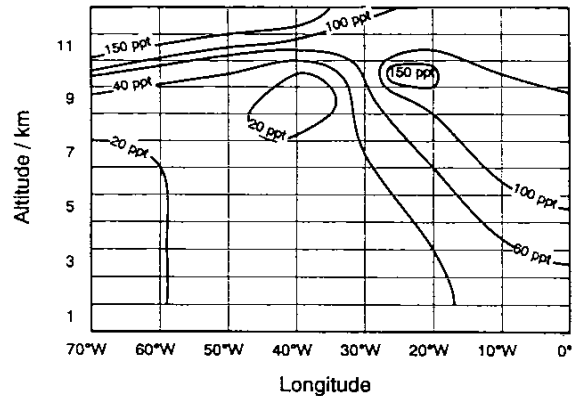


Figure 11-8. Daytime NO mixing ratio distribution (altitude vs. longitude) across the North Atlantic during the period June 4-6, 1984, of the STRATOZ III campaign. (Based on Ehhalt *et al.*, 1993.)

Figure 11-10), Ehhalt *et al.* (1992) could reproduce quite reasonably the measured vertical profiles shown in Figure 11-9. The transport of polluted air masses from the planetary boundary layer to the upper troposphere by fast vertical convection is considered an important process for NO_x by these authors. However, Kasibhatla (1993) suggests that the stratospheric source is a more important source than that arising from rapid vertical convection, but the calculations did not consider lightning, biomass burning, and soil emissions, and the heterogeneous removal of N₂O₅.

Despite considerable differences in model transport characteristics and emission rates, all the studies suggest that aircraft are important contributors to upper tropospheric NO_x and NO_y concentrations. For example Ehhalt *et al.* (1992) suggest that aircraft emissions (estimated for 1984) contribute around 30% to upper tropospheric NO_x (Figure 11-10). Kasibhatla (1993) estimates that about 30% of the NO_x in the upper troposphere between 30° and 60°N are from aircraft. It is clear from the results of Beck *et al.* (1992) and Kasibhatla (1993) that despite large latitudinal variations in the rate of aircraft emissions, the impacts become manifest over the entire zonal band, though not evenly. This behavior is in contrast to the behavior in the lower troposphere, and is due to the slower conversion of NO_x to form HNO₃, and the slower removal rates for HNO₃, which allow for reconversion back to NO_x.

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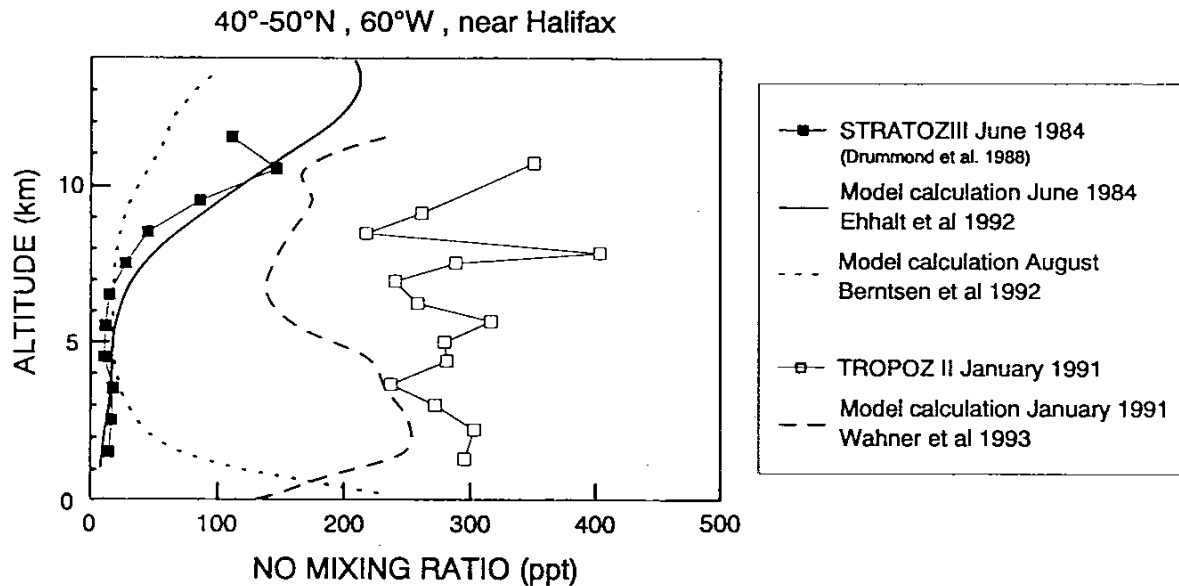


Figure 11-9. Comparisons of measured vertical profiles of NO (June 1984 and January 1991) with calculations from two-dimensional models. (Based on data from: Wahner *et al.*, 1994; Berntsen and Isaksen, 1992; Drummond *et al.*, 1988.)

Several authors discuss the changes to OH concentration consequent to the growth in ozone, and the consequences to methane destruction. Beck *et al.* (1992) predicts OH changes of +10% at around 10 km for the region 30°-60°N. Similar values are suggested by Fuglestedt and Isaksen (1992) (+20%) and Rohrer *et al.* (1993) (+12%). These subsonic aircraft results should be considered as being preliminary given the complexity of the models, the lack of model intercomparison exercises, as well as the paucity of measurements to test against model results.

11.6 CLIMATE EFFECTS

Both subsonic and supersonic aircraft emissions include constituents with the potential to alter the local and global climate. Species important in this respect include water vapor, NO_x (through its impact on O₃), sulfur, soot, cloud condensation nuclei, and CO₂. However, quantitative assessments of the climate effects of aircraft operations are difficult to make at this time, given the uncertainty in the resulting atmospheric

composition changes, as well as uncertainties associated with the climate effects themselves. Therefore, the following discussion will be on possible mechanisms by which aircraft operations might affect climate, along with some estimates of their relative importance.

Increases of CO₂ and water vapor, and alterations of ozone and cirrus clouds have the potential to alter *in situ* and global climate by changing the infrared (greenhouse) opacity of the atmosphere and solar forcing. Sulfuric acid, which results from SO_x emissions, may cool the climate through producing aerosols that give increased scattering of incoming solar radiation, while soot has both longwave and shortwave radiation impacts. The direct radiative impact for the troposphere as a whole is largest for concentration changes in the upper troposphere and lower stratosphere, where the effectiveness is amplified by the colder radiating temperatures. However, the impact (including feedbacks) on surface air temperature may be limited if changes at the tropopause are not effectively transmitted to the surface (see Chapter 8).

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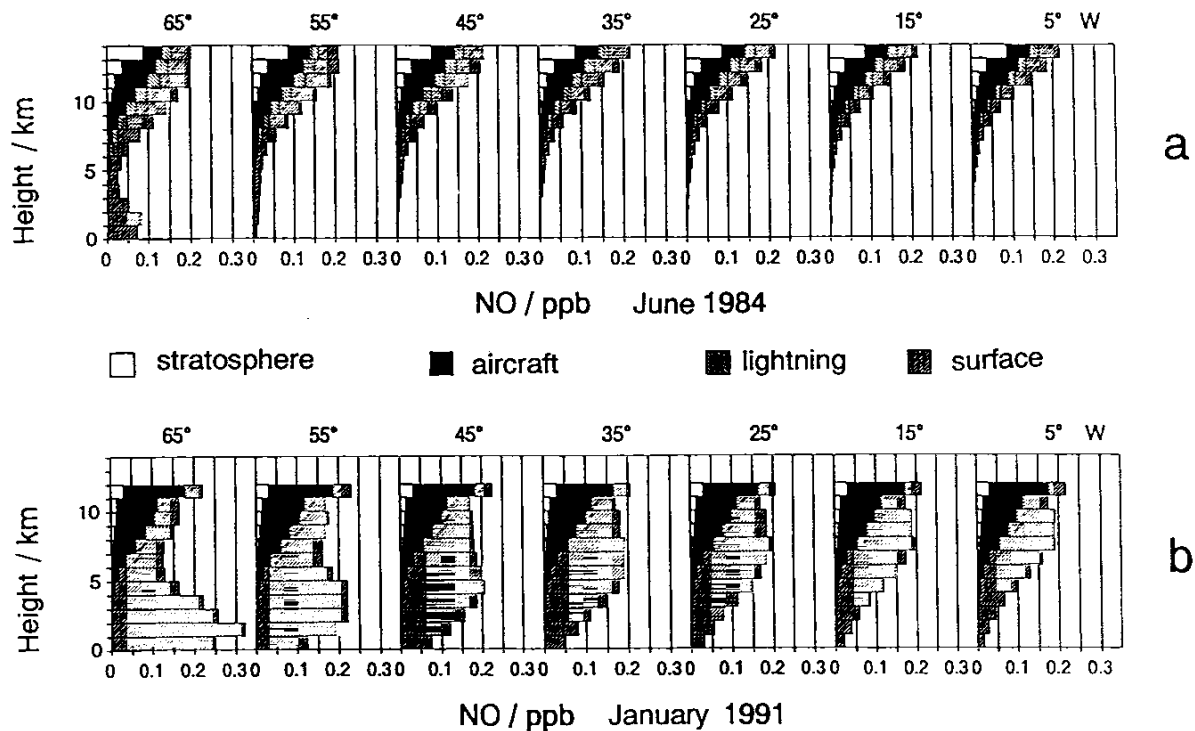


Figure 11-10. Calculations of vertical profiles of NO during summer (June, top panel) and winter (January, bottom panel) using a quasi-two dimensional longitude-height model for the latitude band of 40°-50°N. The different shadings relate to the different sources: stratosphere, lightning, surface (fossil fuel combustion and soil emissions), and aircraft (Ehhalt *et al.*, 1992).

11.6.1 Ozone

As has been discussed in Chapter 8, the impact of ozone changes on the radiation balance of the surface-troposphere system depends on the vertical distribution of the ozone changes. Reduction in tropospheric and lower stratospheric ozone tends to cool the climate, by reducing the atmospheric greenhouse effect. Reduction in middle and upper stratospheric ozone tends to warm the climate, by allowing more shortwave radiation to reach the surface (Lacis *et al.*, 1990).

The preliminary assessments of the HSRP/AESA program are that supersonic aircraft operations could decrease ozone in the lower stratosphere by less than 2 percent for an $EI(NO_x)$ of 15, while increasing it in the upper troposphere by a similar percentage. When these ozone changes were put into the NASA Goddard Institute for Space Studies (GISS) 3-D climate/middle

atmosphere model (Rind *et al.*, 1988), the resulting change in global average surface air temperature was approximately -0.03°C . The net result is a consequence of the net effect of varying influences: ozone reduction in the stratosphere at 20 km, and ozone increases in the upper troposphere produce surface warming, while ozone reduction in the lower stratosphere produces surface cooling. The net result provides the small temperature changes found in this experiment.

Assuming a local ozone increase (8 to 12 km, 30° to 50°N) of 4 - 7% due to doubling of the subsonic aircraft NO_x emission and incorporating these changes into the Wang *et al.* (1991) model, the inference can be drawn that a radiative forcing of 0.04 to 0.07 W m^{-2} will result (Mohnen *et al.*, 1993; Fortuin *et al.*, 1994). This radiative forcing is of the same order as that resulting from the aircraft CO_2 emissions (see Chapter 8.2.1). The estimated feedback on radiative forcing from methane

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decreases (due to the OH increase from increasing NO_x) has been estimated to be small using two-dimensional models (Johnson, 1994; Fuglestedt *et al.*, 1994).

11.6.2 Water Vapor

Water vapor is the primary atmospheric greenhouse gas. Increases in water vapor associated with aircraft emissions have the potential to warm the climate at low tropospheric levels, while cooling at altitudes of release, due to greater thermal emission. The effects are largest when water vapor perturbations occur near the tropopause (Graßl, 1990; Rind and Lacis, 1993), as is likely to be the case.

High-speed aircraft may increase stratospheric water vapor by up to 0.8 ppmv for a corridor at Northern Hemisphere midlatitudes, with a Northern Hemispheric effect perhaps 1/4 as large (Albritton *et al.*, 1993). When changes of this magnitude were used as input to the stratosphere, the GISS climate/middle atmosphere model failed to show any appreciable surface warming, as the radiative effect of the negative feedbacks (primarily cloud cover changes) were as important as the stratospheric water forcing. In general, the stratosphere cooled by a few tenths of a degree, associated with the increased thermal emission.

Subsonic tropospheric emissions of water vapor could possibly result in increases on the order of 0.02 ppmv. Shine and Sinha (1991) estimate that a global increase of 1 ppmv for a 50 mbar slab between 400 and 100 mbar would increase surface air temperature by 0.02°C. Therefore the climate effects from subsonic water vapor emission by aircraft seem to be very small.

11.6.3 Sulfuric Acid Aerosols

Subsonic aircraft, flying both in the troposphere and stratosphere, are presently adding significant amounts of sulfur to the atmosphere. Hofmann (1991) has estimated that the current fleet may be contributing about 65% of the background non-volcanic stratospheric aerosol amount, whose optical thickness is approximately $1 - 2 \times 10^{-3}$; note however, that this view is a controversial one as can be seen in Section 3.2.1 of Chapter 6. This added optical thickness would imply a contribution to the equilibrium surface air temperature cooling on the order of 0.03°C due to aircraft sulfur emissions (Pollack *et al.*, 1993).

11.6.4 Soot

Particles containing elemental carbon are the result of incomplete combustion of carbonaceous fuel. Such particles have greater shortwave absorbing characteristics than do sulfuric acid aerosols, and thus a different shortwave/longwave impact on net radiation. Upper tropospheric aircraft emissions of soot presently account for about 0.3% of the background aerosol (Pueschel *et al.*, 1992).

The total soot source for the stratosphere is currently estimated as 0.001 teragrams/year (Stolarski and Wesoky, 1993b), most likely coming primarily from commercial air traffic. This accounts for about 0.01% of the total stratospheric (background) aerosol loading (Pueschel *et al.*, 1992). It is estimated that the proposed HSCT fleet would double stratospheric soot concentrations for the hemisphere as a whole, while increases of up to a factor of ten could occur in flight corridors (Turco, 1992).

11.6.5 Cloud Condensation Nuclei

Contrails in the upper atmosphere act in a manner somewhat similar to cirrus clouds, with the capability of warming the climate by increasing longwave energy absorption in addition to the shortwave cooling effect. Aircraft sulfur emissions in addition to frozen droplets are the most likely contributor to this "indirect" effect of aerosols, but soot might also be important.

The impact of aircraft particle emissions on upper tropospheric cloud amounts and optical processes is not yet known, though it is likely to grow with increased air traffic. Changes in cloud cover and cloud optical thickness resulting from aircraft operations might be the most significant aircraft/climate effect, but quantitative evaluations of this are very uncertain. In a 2-D analysis, increases in cirrus clouds of 5% between 20-70°N produced a warming of 1°C, due to increased thermal absorption (Liou *et al.*, 1990). For 0.4% additional cloud coverage by contrails and mid-European conditions, an increase in surface temperature of about 0.05°C is estimated (Schumann, 1994).

11.6.6 Carbon Dioxide

While aircraft CO₂ emissions are at a different altitude from other anthropogenic emissions, the climate

impact should be qualitatively similar, as CO₂ is a relatively well-mixed gas. Therefore the climate impact from subsonic CO₂ emissions can be estimated to be approximately 3% of the total anthropogenic CO₂ impact, since subsonic aircraft fuel consumption is about 3% of the global fossil fuel consumption.

11.7 UNCERTAINTIES

This chapter deals with the atmospheric effects of both the present subsonic aircraft fleet and an envisioned future supersonic aircraft fleet. The uncertainties in assessing these two atmospheric effects are of a different nature. For instance, there is a real uncertainty in the present emissions data base that results from uncertainties in the aircraft engine characteristics, engine operations, and air traffic data. There are also uncertainties relating to the models being used to examine the atmospheric effects of these subsonic emissions. In the supersonic case, assessments are being made for a hypothetical aircraft fleet, so modeling uncertainties are the main concern. The modeling uncertainties are probably much greater than the emission uncertainties at the present time.

11.7.1 Emissions Uncertainties

As was indicated previously, the evaluation of a time-dependent emissions data base for use in atmospheric chemical-transport models requires a rather complete knowledge of the specific emissions produced by all types of aircraft, as well as a knowledge of the operations and routing of the aircraft fleet.

There has been very limited aircraft engine testing under realistic cruise conditions for the present subsonic aircraft fleet. At the present time, some engine tests are being carried out under simulated altitude conditions to see if the present method of determining NO_x, for example, from a combination of theoretical studies and laboratory combustor testing can be validated.

A disagreement exists between the quantity of fuel produced and predicted fuel usage by the data bases. This discrepancy probably results from uncertainties in emissions for the non-OECD (Organization for Economic Cooperation and Development) countries and for military traffic, and from the uncertain estimates of loading and power settings of the aircraft fleet.

11.7.2 Modeling Uncertainties

There are two types of modeling uncertainties in the aircraft assessment process. One is related to modeling of small-scale plume processes, while the other relates to the global atmospheric modeling.

PLUME MODELING

As was indicated earlier in this chapter, considerable modeling is required to characterize the evolution of the aircraft exhaust leaving the engines' tailpipes to flight corridor spatial scales and then to the scales that are treated in the atmospheric models of aircraft effects. These plume models must treat turbulent dynamics and both gas phase and heterogeneous chemistry. Only one such model presently exists that treats the full problem and there exists no measurement program that is aimed at the validation of this model (Miake-Lye *et al.*, 1993). There have been very few actual measurements in airplane exhaust wakes. There are the chemical measurements at altitudes of about 10 km by Arnold *et al.* (1992), and there were turbulence and humidity data taken by Baumann *et al.* (1993) at the same time. Also, there are the SPADE (Stratospheric Photochemistry, Aerosols, and Dynamics Expedition) measurements taken during crossings of the ER-2 exhaust plume (Fahey *et al.*, 1994). These measurements, while valuable, are not sufficient to validate the plume processing model.

ATMOSPHERIC MODELING

The upper troposphere and lower stratosphere, the regions of major interest in this chapter, are particularly difficult regions to model. In 2-D models of supersonic aircraft effects, the meridional transport circulation is difficult to obtain since the radiative heating is comprised of a number of small terms of different sign. Thus, small changes in any radiation term can have important consequences for transport. Similarly, the time scales for both transport and chemistry to modify the ozone distribution are generally long and comparable. The complete problem must be solved. The NO_x, HO_x, and ClO_x chemical processes are highly coupled in the stratosphere. Modeling the chemical balance correctly, in regions where few measurements are available, presents formidable difficulties. This situation is even worse in the upper troposphere than in the stratosphere, given that

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the chemistry of the upper troposphere is more complex and there are fewer existing observations of this region.

Supersonic aircraft have their cruising altitudes in the middle stratosphere (near 20 km) while subsonic aircraft have cruise altitudes that lie both in the troposphere and lower stratosphere. Supersonic assessment calculations have been done using 2-D models up to the present time, while it is generally appreciated that 3-D models will be necessary for credible subsonic assessments. Thus, separate discussions of modeling uncertainties follow for aircraft perturbations in the stratosphere and in the troposphere.

TRANSPORT

Two particular problems relating to atmospheric transport are extremely important for the supersonic aircraft problem. First, stratosphere-troposphere exchange, which cannot be modeled in detail with great confidence in global (2-D or 3-D) models, is clearly of special significance to the chemical distribution in these regions, to the lifetime of emitted species, etc. More work on this topic is essential. Second, the present 2-D assessment models do not model well the details of the polar vortex, although improvements are anticipated when these models include the Garcia (1991) parameterization for breaking planetary waves. If the ideas of the polar vortex as a "flowing processor" are correct (see Chapter 3), then the correct modeling of polar vortex dynamics will have a crucial impact on the distribution of species in the lower stratosphere, and present 2-D models would clearly be performing poorly there. There is also the larger issue that the uncertainty connected with the use of 2-D models to assess the inherently 3-D aircraft emission problem needs to be evaluated further. Even when 3-D models are available to model this problem, however, the question will remain as to how well these 3-D models simulate the actual atmosphere until adequate measurement-model comparisons are done.

For modeling aimed at assessing the atmospheric effects of both subsonic and supersonic aircraft, it is crucial to properly model ambient NO_x distributions in the upper troposphere, and these, in turn, depend on properly modeling transport between the boundary layer and the free troposphere, on proper modeling of the fast upward vertical transport accompanying convection, and on modeling the lightning source for NO_x . Considerable effort is needed to improve our capability in these areas.

It is also necessary to model stratospheric-tropospheric transport processes carefully so that NO_x fluxes and concentrations in the region near the tropopause are realistic. This requires a substantial effort to improve our understanding of stratosphere-troposphere exchange processes.

CHEMICAL CHANGES

The effect of NO_x emitted by subsonic aircraft depends on the amount of NO_x in the free troposphere. The ambient NO_x concentrations are not very well known, and depend on several factors such as surface emission from anthropogenic and natural biogenic sources, the strength of the lightning source for NO_x , and the transport of stratospheric NO_x into the troposphere (see Chapter 2, Table 2-5). The inclusion of wet and dry deposition processes and entrainment in clouds in assessment models is at a very preliminary stage.

Heterogeneous chemistry is another important area of uncertainty for models of the troposphere and lower stratosphere. For example, the hydrolysis of N_2O_5 is important in both the troposphere and stratosphere, but the precise rate for this reaction is not known. Observational studies are needed to elucidate the exact nature and area of the reactive surfaces. Furthermore, at the present time, heterogeneous chemistry is being crudely modeled. Although there do exist models describing the size distribution and composition of stratospheric aerosols, no aircraft assessment model presently exists that incorporates and calculates aerosol chemistry.

In supersonic assessment models, it is important to properly model the switch over (at some altitude) from NO_x -induced net ozone production to net ozone destruction. The precise altitude at which this switch over occurs differs from model to model, and this can lead to very different ozone changes in different models of supersonic aircraft effects. The different responses of the various models used in the HSCT/AESA assessment of the impact of changed EI (see Tables 11-4 and 11-5, for example) point to important, unresolved differences in these models that must be addressed before a satisfactory assessment of the atmospheric effects of supersonic aircraft can be made with confidence. Also, it is clear from examining the modeled O_3 changes in Chapter 6 that the model results at altitudes below about 30 km differ significantly from one another. They also do not give as large O_3 losses as are observed (see Chapter 1). This

problem is particularly acute if one accepts the SAGE results indicating large decreases in ozone concentrations just above the tropopause (see Chapter 1) as being correct. Then, the fact that present stratospheric models do not correctly give this effect casts doubt on present assessment models to correctly simulate that atmospheric region. Since it is in this region where effects from aircraft operations are particularly significant, there is the question of how well we can correctly predict atmospheric effects in this altitude region. It may be that the SAGE ozone trends in this region are in error, or it may be that important effects in this region are not properly included in present models.

11.7.3 Climate Uncertainties

The study of the possible impact of aircraft on climate is now just beginning. One can make some preliminary extrapolations based on existing climate research, but one should appreciate that the complexity of climate research, in general, implies that it will be some time before great confidence can exist in estimates of aircraft impacts on climate.

11.7.4 Surprises

Early assessments of the impact of aircraft on the stratosphere varied enormously with time as understanding slowly improved. Our understanding of the lower stratosphere/upper troposphere region is still far from complete and surprises can still be anticipated, which may either result in greater or lesser aircraft effects on the atmosphere.

ACRONYMS

| | |
|---------|---|
| AER | Atmospheric and Environmental Research, Inc. |
| AERONOX | The Impact of NO _x Emissions from Aircraft upon the Atmosphere |
| AESA | Atmospheric Effects of Stratospheric Aircraft |
| ANCAT | Abatement of Nuisance Caused by Air Traffic |
| CAMED | University of Cambridge and University of Edingburgh |
| CEC | Commission of the European Communities |
| CIAP | Climatic Impact Assessment Program |
| ECAC | European Civil Aviation Conference |
| ECMWF | European Centre for Medium-Range Weather Forecasts |
| EI | Emission Index |
| GISS | NASA Goddard Institute for Space Studies |
| GSFC | NASA Goddard Space Flight Center |
| HSCT | High-Speed Civil Transport |
| HSRP | High Speed Research Program |
| ICAO | International Civil Aviation Organization |
| IEA | International Energy Agency |
| LLNL | Lawrence Livermore National Laboratory |
| LTO | Landing/Take-Off cycle |
| MOZAIC | Measurement of Ozone on Airbus In-service Aircraft |
| NASA | National Aeronautics and Space Administration |
| NCAR | National Center for Atmospheric Research |
| NRC | National Research Council |
| OECD | Organization for Economic Cooperation and Development |
| OSLO | University of Oslo |
| POLINAT | Pollution from Aircraft Emissions in the North Atlantic Flight Corridor |
| SAGE | Stratospheric Aerosol and Gas Experiment |
| SBUV | Solar Backscatter Ultraviolet spectrometer |
| SPADE | Stratospheric Photochemistry, Aerosols, and Dynamics Expedition |
| WMO | World Meteorological Organization |

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number (Tavoularis and Karnik 1989). Hence, turbulent mixing may occur under nonstationary conditions at all Richardson numbers.

When comparing mixing properties in the atmosphere and in the ocean, one has to note the rather large molecular Schmidt number of salt diffusing in water (about 500) while the corresponding Prandtl number of thermal diffusion in air is about 0.7. At high Reynolds numbers, one generally expects that the large-scale turbulent motions become independent of the Prandtl number, at least for neutral stratification. However, for strong stratification, Pearson et al. (1983) show that the vertical diffusivity is limited by small-scale mixing once the available kinetic energy is consumed to provide the potential energy required for vertical displacements. Such small-scale processes will depend on molecular diffusion.

The present theory uses several assumptions in order to allow for a simple analytical model. It assumes that the density variations affect the buoyancy only, that is, we employ the Boussinesq approximation. The analysis is restricted to flows at high Reynolds numbers with active turbulence and with small molecular diffusion at the energy containing scales. The turbulence is further assumed to be strongly sheared ($Ri < 1$) so that the timescale S^{-1} of shear is smaller than the timescale N^{-1} of stratification, and both should be smaller than the turbulence timescales. The theory assumes a homogeneously turbulent flow exposed to uniform (linear) vertical shear and stratification. Because of homogeneity, all divergences of fluxes vanish so that also the mean profiles do not change due to turbulent mixing. As a result, the mean flow is characterized by a unique value of Ri . The assumption of nearly homogeneous turbulence is appropriate when the length scales of turbulence are small compared to outer scales of any variations in the mean profile such that the divergence of fluxes is small compared to the local rate of energy dissipation. By means of scale analysis, Mellor and Yamada (1974) have shown that this assumption of nearly homogeneous turbulence is often satisfied even in inhomogeneous boundary layers. Finally, we assume that the exchange of energy between its kinetic and potential form has approached a local equilibrium so that averaged quantities decay in a nonoscillating manner. Hence, the model excludes situations with large amplitude wavy oscillations between kinetic and potential forms of energy.

Homogeneous turbulence is by necessity time dependent and becomes stationary only under special conditions near a "stationary" Richardson number. Certainly, the transient states at other Richardson numbers cannot last forever. For stable stratification the energy will decay and disappear until any event, which the present model will not explain, creates a new turbulent region. The model loses validity when the turbulence gets so weak that viscosity becomes important. For growing turbulence in a large but finite

domain, the assumption of homogeneity eventually breaks down when growing spatial gradients cause substantial energy diffusion out of the finite turbulent region.

Homogeneous turbulent shear flows have been measured by Rohr et al. (1988) in salt-stratified water. Their data are taken from appendix 2 of Rohr (1985) at shear times $tdU/dz > 6$ when the flow has approached constant correlation coefficients. Reliable data for homogeneous air flows are available only for neutral stratification (Tavoularis and Karnik 1989). In order to extend the database, we use results from numerical simulations of three-dimensional turbulence in homogeneous stratified shear flows at a Prandtl number of one.

Homogeneous turbulence in stratified shear flows has been investigated by direct numerical simulation (DNS) in Gerz et al. (1989), Gerz and Schumann (1991), and Holt et al. (1992). They investigated the flow dynamics as a function of Richardson numbers in between 0 and 1.32. DNS on grids with 128^3 grid points, as we could realize for this study, is typically restricted to a Prandtl number of order unity and to a turbulent Reynolds number, based on root-mean-square velocity fluctuations and Taylor's microscale, of less than about 50. For atmospheric flows, much larger Reynolds numbers are of interest. For this reason, the DNS method has been extended by Kaltenbach (1992) into a large-eddy simulation (LES) using a simple subgrid-scale model (Lilly 1967). This extends formally the Reynolds number to infinity. However, the range of resolved scales is still limited by numerical resolution. The method and the parameters used for LES are summarized in the appendix. Details and further results are reported in Kaltenbach et al. (1994). These are the first LES of stably stratified sheared homogeneous turbulence. LES of turbulence in the stable atmospheric boundary layer have been performed by Mason and Derbyshire (1990), and these results have been used by Derbyshire and Hunt (1993) to investigate mixing models.

The paper is organized as follows. In section 2, a simple theory is deduced based on the budget of kinetic energy. Here, temperature fluctuations are assumed to be correlated with shear for small Richardson numbers and with stratification for large Richardson numbers. The results of this model are compared to several experiments and to the results of the LES. Section 3 extends the model using the budget of potential energy to determine the dependence of temperature fluctuations on Richardson number. Section 4 discusses various assumptions and consequences of the model and shows that the results are not far from measurements in the stationary but inhomogeneous boundary layer and in some stratified flows with weak shear. Finally, section 5 summarizes the conclusions.