Assessing the Impact of Aircraft Emissions on the Stratosphere

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Background

Research into the impacts of aircraft exhaust on stratospheric chemistry and climate began with the Climate Impact Assessment Program (CIAP) in the 1970s, well before chlorofluorocarbons and CO_2 climate warming became prominent. The scientific issues associated with assessing the impact of aircraft are central to understanding stratospheric chemical, dynamical, radiative, and microphysical processes. For the past decade, the NASA Atmospheric Effects of Aviation Project (AEAP) has been the U.S. focal point for research on aircraft effects. In conjunction with U.S. basic research programs, AEAP and concurrent European research programs have driven remarkable progress in understanding the atmospheric effects of aviation, culminating in two major assessment reports released in 1999 [IPCC, 1999; Kawa et al., 1999]. The former report primarily focuses on aircraft effects in the upper troposphere, with some discussion on stratospheric impacts. The latter report focuses entirely on the stratosphere. The current status of research regarding aviation effects on stratospheric ozone and climate, as embodied by the findings of these reports, is reviewed here.

Two main classes of aircraft are important to consider for the current and future stratosphere. The first is the existing commercial fleet that flies at subsonic speeds in the altitude region around 11 km. These flights take place in the lower stratosphere about 20% of the time. The commercial fleet is projected to grow by approximately a factor of 4 in the next 50 years. The second class of aircraft is the proposed, hypothetical fleet of high-speed civil transport (HSCT) aircraft that would cruise at supersonic speeds (Mach 2.4, 1600 mph) near 19-km altitude, the middle of the stratospheric ozone layer. The HSCT follows on the supersonic transport (SST) aircraft first discussed in the 1970s [Johnston, 1971]. Recently the HSCT development effort, supported by NASA and the aerospace industry [Wilhite and Shaw, 1997], has been suspended. Thus the deployment of a substantial HSCT fleet will probably not occur in the next 20 to 30 years. The method, problems, and findings of the HSCT assessment, however, are relevant to numerous fundamental and applied problems in stratospheric research.

The aircraft emissions of primary concern for stratospheric ozone and climate are oxides of nitrogen (NO_x), water (H_2O), and aerosol particles and particle precursor gases. Unburned hydrocarbons, carbon monoxide, and soot emissions are also considered but their impact in the stratosphere is generally considered negligible and will not be further discussed.

Nitrogen oxides are involved in the principal loss process for ozone in the middle and upper stratosphere, and thus, exhaust that is transported to these regions will reduce ozone. In the lower stratosphere, NO_x radicals moderate ozone loss due to other radical species (hydrogen oxides (HO_x), chlorine oxides (ClO_x), bromine oxides (BrO_x)); thus addition of NO_x from aircraft exhaust can either increase or decrease ozone in this region depending on the relative balance among the radicals. Increasing NO_x in the upper troposphere leads to increased ozone. In the

polar winter stratosphere, nitrogen oxides participate in formation of polar stratospheric clouds (PSCs), which lead to large seasonal ozone loss, e.g., the Antarctic ozone "hole." The net effect of increasing NO_x depends on interactions between transport, heterogeneous chemistry, homogeneous chemistry, and the composition of the unperturbed atmosphere.

Future HSCT emissions could increase lower stratospheric water vapor by about 0.5 ppmv (10 to 15% for a fleet of 500 aircraft) affecting climate, aerosol processes, and rates for chemical reactions. Warming of the lower atmosphere as a result of increased stratospheric water is predicted to be the main climatic effect of HSCTs. Since water is the source of HO_x radicals, increased water leads directly to higher concentrations of HO_x. The composition and growth of aerosol particles, including PSCs, is influenced because increased water vapor raises the condensation temperature. Over most of the stratosphere, however, the temperature is too warm and humidity too dry for the emission of water from aircraft to produce persistent clouds. Increased water also increases the reactivity of aerosol toward gases, such as HCl and $ClONO_2$, thus influencing the relative concentrations of radical species. Model calculations suggest that the associated increase in HO_x is as important as changing NO_x in determining ozone change.

Repeated observations since 1994 consistently show that a large number of ultrafine (<20 nm diameter) aerosol particles exist in jet engine exhaust plumes, and that particle production increases as the sulfur content of fuel increases. Emission of small particles and sulfur dioxide (SO_2) can potentially increase aerosol surface area throughout the stratosphere, which suppresses NO_x and enhances ozone loss by ClO_x and HO_x. Proposed mechanisms for small particle formation are still controversial, and the effects on particle abundance throughout the stratosphere are uncertain, but atmospheric ozone is definitely sensitive to changing aerosol conditions [WMO, 1999]. In the upper troposphere aircraft particle production is a major concern because it may lead to changes in cirrus cloud properties and associated cloud radiative forcing (see IPCC [1999]).

The key factors that determine the atmospheric impacts of aircraft emissions are:

- The quantity of exhaust deposited (water, NO_x, particle mass and surface area) and its location in altitude and latitude;
- Atmospheric transport, especially the eventual accumulation of exhaust products in various parts of the stratosphere;
- Chemical reactions of the exhaust products with aerosols, atmospheric radicals, and ozone;
- Microphysics (formation, growth, coagulation, and settling) of aerosol particles in the atmosphere; and
- The background state (meteorology and composition) of the atmosphere onto which the aircraft perturbation is superimposed.

Because the impact of the current fleet is not discernable from other sources of variability in the stratosphere and the future fleet does not exist yet, we must simulate these processes and

estimate the impacts in numerical models. Much of the uncertainty in the results derives from uncertainty in atmospheric modeling. Progress and uncertainties associated with key issues in calculating the impact of aircraft in the stratosphere are discussed below. Most of the discussion pertains to the effect of emission by HSCTs directly into the stratosphere. The impacts of subsonic aircraft in the stratosphere are also briefly discussed.

Aircraft Emissions

The three-dimensional (3-D) deposition of aircraft exhaust into the atmosphere is known to relatively good accuracy, within about 20% [IPCC, 1999]. Projections of future fuel use and exhaust deposition are more uncertain, and the impact of varying assumptions for the future (e.g., number of aircraft deployed and timing of introduction into service) is tested parametrically by running the models with a variety of different scenarios (e.g., Figure 1). The specific exhaust emissions are expressed as an emission index (EI) in grams of emitted species per kilogram of fuel used. Current engine EIs are well known, and the gas-phase chemistry taking place in the aircraft near-field plume are generally confirmed by in situ measurements with one notable exception.

More small volatile particles are formed in jet aircraft exhaust than previously expected, and the mechanism and control of this production are currently not well understood. Important progress has been made with new direct measurements for existing aircraft that show formation of volatile ultra-fine aerosol particles in exhaust plumes from all aircraft sampled. In-flight measurements indicate that the number of particles is dependent on fuel sulfur content, while altitude chamber measurements show that sulfur emissions at the engine exit plane are primarily SO_2. These observations support earlier inferences of a composition of sulfuric acid (H_2SO_4)/H_2O for the volatile particles detected in the plume, but the particle emission yield for the HSCT is still very uncertain. The atmospheric effect depends on the fraction of the emitted sulfur that is in particle phase versus gas since the SO_2 gas is oxidized after mixing with ambient air and deposited on existing particles giving a smaller increment to total sulfate particle surface area. Model calculations testing the atmospheric sensitivity to a range of particle emissions under differing atmospheric aerosol loading, which are mainly controlled by volcanic eruptions, result in a range of impacts larger than that attributed to nitrogen oxides or water.

Transport

The effect of aircraft exhaust depends strongly on its accumulation and dispersion within the stratosphere. The exhaust distribution depends on the aggregate over many different transport processes; in particular, transport from mid-latitude flight corridors into the tropics, strength of meridional circulation, and transport out of the stratosphere into the troposphere. These same processes also determine the distribution of source gases in the background atmosphere. Concern about transport arises from simulations of the current atmosphere. To the extent that the model distributions of tracers and ozone do not match reality, the aircraft perturbation will be superimposed on an incorrect background atmosphere. Moreover, the transported distribution of the aircraft exhaust may not be correct. Models used in the HSCT assessments show a large range (greater than a factor of two) in the peak accumulation and dispersion of the HSCT exhaust

(Figure 2). An even greater difference among models is seen for the distribution of subsonic exhaust in the stratosphere (below).

Observations have begun to constrain several key components of transport necessary to predicting the distribution of aircraft exhaust. *In situ* measurements of chemical tracers have been obtained within the previously data-sparse tropics. These observations permit quantitative diagnosis of key pathways for dispersal of HSCT exhaust into the upper stratosphere where chemical sensitivity to NO_x is high. Measurements of CO_2, SF_6, and HF over a range of latitude and altitude have enabled mean ages of air in the stratosphere to be determined. Age of air is a directly measured diagnostic related to stratospheric residence time and hence to the potential accumulation of HSCT exhaust in the stratosphere. The quantitative analysis of tropical transport and mean age provides stringent new tests of transport within numerical models. Figure 3 shows that models significantly underestimate mean age in the lower stratosphere, suggesting that their stratospheric residence time is too short. On the other hand, model comparisons with measured NO_y (a long-lived tracer in the lower stratosphere) profiles are distributed both higher and lower than the observations, although both background and aircraft delta-NO_y amounts are correlated among models with their calculated age as generally expected. Thus it remains difficult to determine which model transport and aircraft exhaust accumulation is most accurate.

One approach to resolving these uncertainties has been use of 3-D atmospheric models for the first time in the HSCT assessment. Three-dimensional models incorporate a more physically realistic representation of the atmosphere than 2-D models. The modular design of the AEAP Global Modeling Initiative 3-D model has made it possible to test the different components of the model (e.g., the numerical transport algorithm and the source of the wind and temperature fields). Objective criteria for performance with respect to data have been applied. Thus, we discern differences among models in their response to the HSCT perturbation and begin to weigh their results. A major model-measurement comparison and model intercomparison (M&M II) has been conducted [Park et al., 1999], and all models in the AEAP assessment have been tested in comparison to a standard set of performance benchmarks. Also, the 2-D models have incorporated more complete process representations including those for aircraft aerosol exhaust, PSCs, heterogeneous reaction rates, and wave-driven mixing. These model developments give us more confidence in our physical representation of the stratospheric system.

Chemistry

The local response of ozone to changes in NO_x, H_2O, and aerosol is becoming increasingly well understood. Through a combination of laboratory experiments, observations of atmospheric radicals and reservoir species (including the first in summer polar regions), and improved approaches to interpreting these observations, uncertainties in chemistry have been reduced. This establishes confidence that we are not missing significant reactions or unknown species that would alter the calculated response of the chemical system to the aircraft perturbation. Kinetic parameters controlling radical abundances have been constrained from simultaneous observations of radicals from all three major chemical families. For current atmospheric conditions, increases in NO_x will decrease local ozone in the mid to upper stratosphere. However, in the lower stratosphere due to the buffering effect of competing catalytic chemical

cycles, the ozone response is only weakly coupled to NO_x over the range (factor of 2) of NO_x concentrations currently present in lower stratosphere (Figure 4). Variations in the background stratospheric aerosol, NO_x, HO_x, halocarbons, and temperature resulting from natural processes (e.g., volcanic eruptions), changes in industrial activity (e.g., N_2O emissions from fertilizer use), and from changes to climate will affect the response of ozone to aircraft exhaust. Predictions of the effects of HSCT exhaust are particularly sensitive to the abundance of NO_x in the lower stratosphere.

Although improved, the ozone loss chemistry is still not completely resolved in models. Recent measurements suggest inaccuracies in the chemical kinetic rates used in current model calculations of the partitioning of reactive nitrogen between NO_x radical and non-radical species [Gao et al., 1999]. In general, models using current rates predict lower concentrations of radicals than observed, a tendency that would underestimate reductions in ozone; the calculated difference for HSCTs using updated rates is small however. Also, changes in the total ozone column due to HSCT exhaust result from a balance between ozone increases in the aerosol-rich lower stratosphere and ozone losses in the NO_x-rich middle and upper stratosphere. Models differ in the magnitude of the vertical and latitudinal contributions to this critical balance. Continued observations are needed to better resolve chemical processes in the stratosphere.

Polar Processes

Properly predicting the interaction of aircraft exhaust species with cold polar processes is an important component of the aircraft assessment. The heterogeneous processes that take place in the cold temperatures of polar winter are highly non-linear in their dependence on aircraft emitted species, NO_y, H_2O, and sulfate, making this is a highly sensitive regime. As a result of a combination of non-linear reaction processes, phase change transitions, and exponential dependence on the particle size distribution, ozone loss can be highly leveraged by relatively small changes in condensibles at temperatures near those commonly observed in the polar stratosphere. This raises the possibility that synergistic effects may occur among the emitted species increasing the likelihood of severe ozone depletion in the Northern hemisphere polar region. Strong local effects at high latitudes are possible and the impact may be felt at mid-latitudes. Model column ozone losses due to HSCTs are largest at high latitudes in almost all cases. For some models the maximum HSCT ozone loss occurs in polar winter/spring, but the difference among models is large lending little confidence to the quantitative estimates.

Fundamental questions about the microphysics and composition of PSCs limit our ability to parameterize key processes such as sedimentation and heterogeneous chlorine activation, which control winter/spring polar ozone loss. The representation of these processes in assessment models is highly simplified and very model-dependent. Furthermore, the 2-D models used in the aircraft assessment do not properly isolate the winter polar vortex air mass. Lack of isolation of the vortex may lead to too much of the exhaust being transported into the vortex and too much of the processed air being transported out of the vortex. Uncertainties in the microphysics of PSCs, winter polar transport, reactive chlorine activation, and chemical ozone loss will all be addressed directly in the upcoming SAGE III Ozone Loss and Validation Experiment (SOLVE) scheduled for Northern hemisphere winter, 1999-2000 (see http://cloud1.arc.nasa.gov/solve/).

Climate Impacts of Supersonic Aircraft

The climate forcing attributable to an HSCT fleet in the year 2050 is predicted to result in a warming which is small relative to that expected from other anthropogenic sources. The total radiative forcing from 1000 HSCTs is estimated to be +0.1 W m^{-2} in 2050, mostly resulting from increased water in the lower stratosphere. Heating from increased water is offset slightly by cooling from decreased ozone. This HSCT number is a concern because the radiative forcing is disproportionately large for the amount of fuel used and equivalent to about 50% of the forcing from the entire projected subsonic fleet. Climate forcing is sensitive to HSCT emissions because the H_2O accumulation is localized in the lower stratosphere. The uncertainty in the HSCT climate forcing is estimated to be about a factor of 3 due to uncertainty in the exhaust accumulation and uncertainty in the temperature adjustment to a non-uniform perturbation of radiatively active gases in the stratosphere [IPCC, 1999].

Subsonic Aircraft Effect on the Stratosphere

Subsonic aircraft in the altitude range 9-13 km fly about 80% and 20% of the time in the troposphere and stratosphere, respectively [Gettelman and Baughcum, 1999]. However, on some routes aircraft spend considerably more time in the stratosphere. For example, in the North Atlantic flight corridor in winter, as much as 65% of flights occur in the stratosphere. Virtually all of the emissions into the troposphere react locally and/or are scavenged from the troposphere to the surface on time scales of a few days. The emissions into the stratosphere occur predominantly at northern mid to high latitudes with a greater amount of deposition during the winter. The portion of the stratosphere accessible to subsonic aircraft is a region where mixing between the troposphere and stratosphere may occur along isentropic surfaces, the stratospheric "middle world" [Holton et al., 1995]. In this region, emissions deposited into the stratosphere have a relatively high probability of mixing into the troposphere. Recently Danilin et al. [1998] studied the potential effects of subsonic aircraft on stratospheric ozone and the residence time of emitted species in the stratosphere. Results from 2-D and 3-D global models varied in their prediction of stratospheric residence time from about 20 to 60 days. They concluded that individual model treatment of numerical diffusion was a likely source of differences. Schoeberl and Morris [1999] have studied transport of aircraft emissions utilizing a parcel trajectory model to estimate residence time of subsonic aircraft emissions in the stratosphere. They conclude that subsonic emissions have a residence time of less than three months. Because of the diabatic downward motion in the stratosphere at mid to high latitudes, very little subsonic exhaust emissions persist in the stratosphere and the impact on ozone loss is small. Emissions from HSCT aircraft, which are deposited at higher altitudes (see Figure 1), have residence times in excess of a year. Testing the validity of these models is difficult, however, and the IPCC [1999] report concludes that uncertainties are still large, due to the lack of model resolution near the tropopause and the related issue of proper depiction of stratosphere-troposphere exchange.

Calculations of the Supersonic Impact on Ozone

Based on a combination of model calculations and expert judgement, the estimated column ozone change in the Northern Hemisphere is -0.4% for a fleet of 500 HSCTs flying Mach 2.4 with an EI_{NO_x} of 5 g/kg, EI_{SO_2} of 0.4 g/kg, and 10% of fuel sulfur converted to particles.

Including the uncertainty in component processes, the hemispheric ozone response is estimated to be in the range of -2.5 to +0.5% and –3.5 to +1.0%, respectively, in the AEAP and IPCC reports. We note that the maximum seasonal and latitudinal ozone changes will be greater than the hemispheric annual mean. Calculations indicate that these ozone changes will lead to increases in exposure to ultraviolet irradiance at the ground in mid latitudes of approximately 0.5%.

Sensitivity to Input Conditions

The sensitivity of the aircraft impact to varying assumptions about emissions (e.g., fleet size, EI, altitude of emissions) and the future atmosphere has been tested in parametric model studies over a range of scenarios. Several findings relevant to HSCT design issues come out of the atmospheric assessment.

The HSCT impact on ozone depends directly on total emissions, i.e., fleet size and fuel use.

Water vapor, which is inherent to jet fuel combustion, accounts for a major part of the calculated stratospheric ozone impact. Increased water vapor in the stratosphere may also contribute to global climate warming.

NO_x emissions are important. Although current atmospheric models do not show much relative sensitivity to very low (EI_{NO_x} = 5 to 10) emissions, higher NO_x emissions clearly increase the impact, especially for larger fleet sizes.

Production of sulfate aerosol particles makes a significant contribution to the calculated ozone impact. This implies that low-sulfur fuel options and methods to control production of particle precursors should be explored.

Flying the HSCT at lower altitudes reduces stratospheric impacts. The atmospheric residence time of the exhaust is decreased and the chemical sensitivity is reduced.

Special issues are associated with exhaust build-up in polar regions, both winter and summer. Under current HSCT route scenarios, direct emissions into the polar vortex are minimal.

Aircraft in the future would operate in a stratosphere that will likely have different trace constituent mixing ratios and aerosol abundances. Climate change from increasing CO_2 will also change stratospheric temperatures and winds. Future changes in these and related quantities cannot be predicted with high accuracy. Assessment calculations using a range of input future conditions have not identified any particular sensitivities different from the standard projections [Kawa et al., 1999], but the applicability to future conditions is less certain. Calculations with varying aerosol background show a decreased sensitivity to aircraft in volcanically enhanced conditions.

Summary

The assessment results provide guidance for informed decisions on environmental policy and aviation technology development, and they provide direction for future stratospheric research. Great progress in stratospheric science in the last decade has been spurred by the focus on assessing aviation impacts. The pathway to future progress in understanding stratospheric processes and their role in climate lies closely in line with that for reducing the uncertainty of aircraft impacts on the stratosphere. This progress will be achieved through continued investment in observations coupled with advances in modeling the stratosphere/troposphere system.

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Figure Captions

Figure 1. Annual average aircraft NOx emissions as a function of altitude and latitude for 1992 and 2015 from the NASA AEAP data base. The emissions for 2015 assume a fleet of 500 HSCTs operating at EI(NOx) = 5 g(NO_2)/kg(fuel). The range of monthly zonal mean tropopause heights for 1993 are superimposed as solid black lines (from National Centers for Environmental Prediction data).

Figure 2. Calculated HSCT-induced change in NO_y (ppbv) during June from the models participating in the AEAP assessment. Results are shown for the scenario with EI_{NO_x} = 5, 500 aircraft, relative to the subsonic only condition. Contours are drawn for 0.0, 0.1, and 0.2 ppbv, and in increments of 0.2 ppbv thereafter.

Figure 3. Comparison of mean ages of air from observations and models for a latitude profile at 20 km and vertical profiles in the tropics, midlatitudes, and high latitudes. The shaded region indicates the range of mean ages from a majority of models in the M&M II comparison [Park et al., 1999] while the curves (without symbols) correspond to mean age profiles from the GSFC (dashed) and Monash1 (solid) models. The symbols correspond to mean age inferred from observations: *in situ* CO_2 (triangles), *in situ* SF_6 (diamonds), and whole-air samples of SF_6 (asterisk, pluses). Adapted from Hall et al. [1999].

Figure 4. Schematic representation of the response of stratospheric ozone loss to changing NO_x, assuming fixed Br_y, Cl_y, and OH (adapted from Wennberg et al. [1994]).

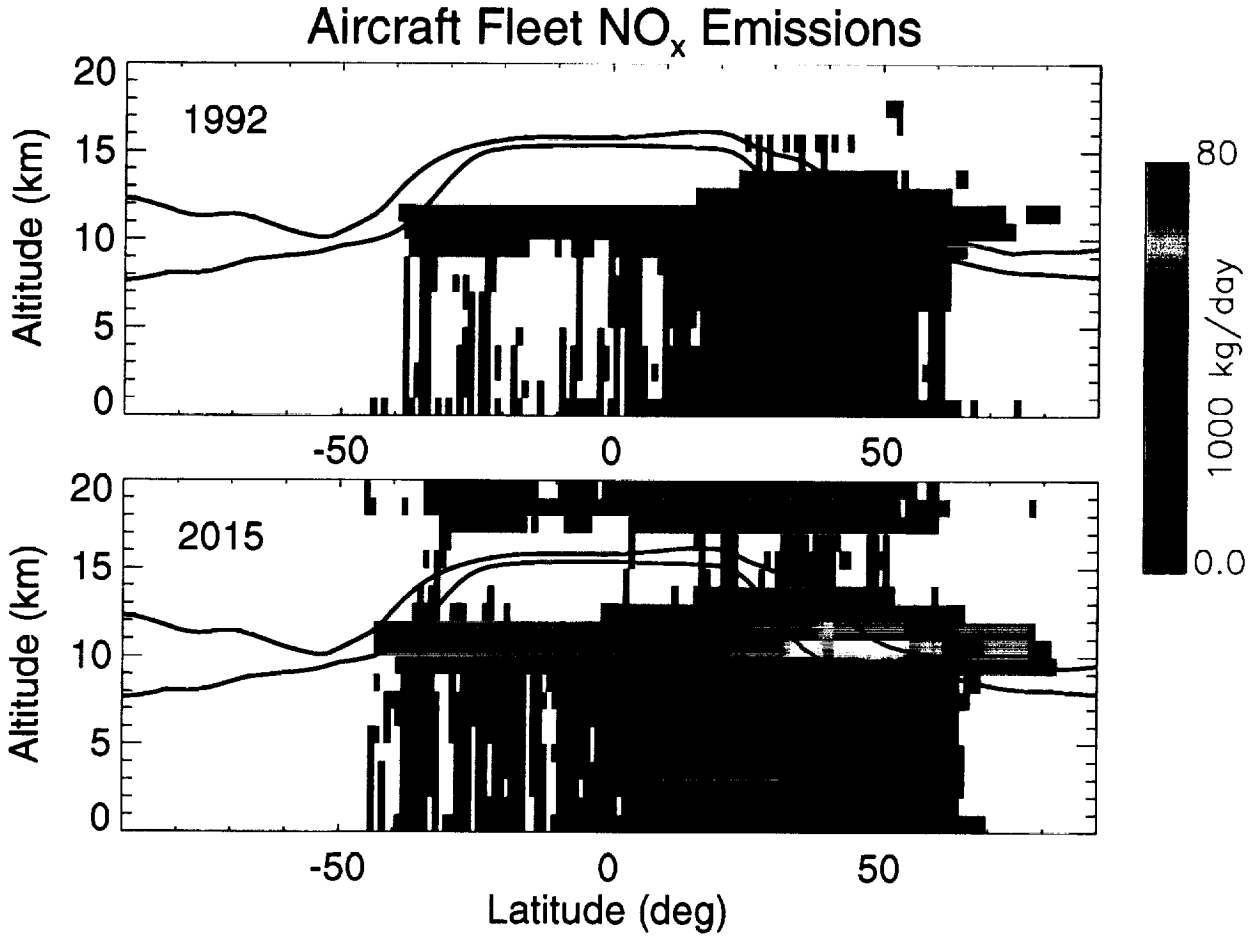
Aircraft Fleet NO_x Emissions
1992
2015
Altitude (km)
20
15
10
5
0
-50
0
50
Latitude (deg)
80
1000 kg/day
0.0

Fig. 1

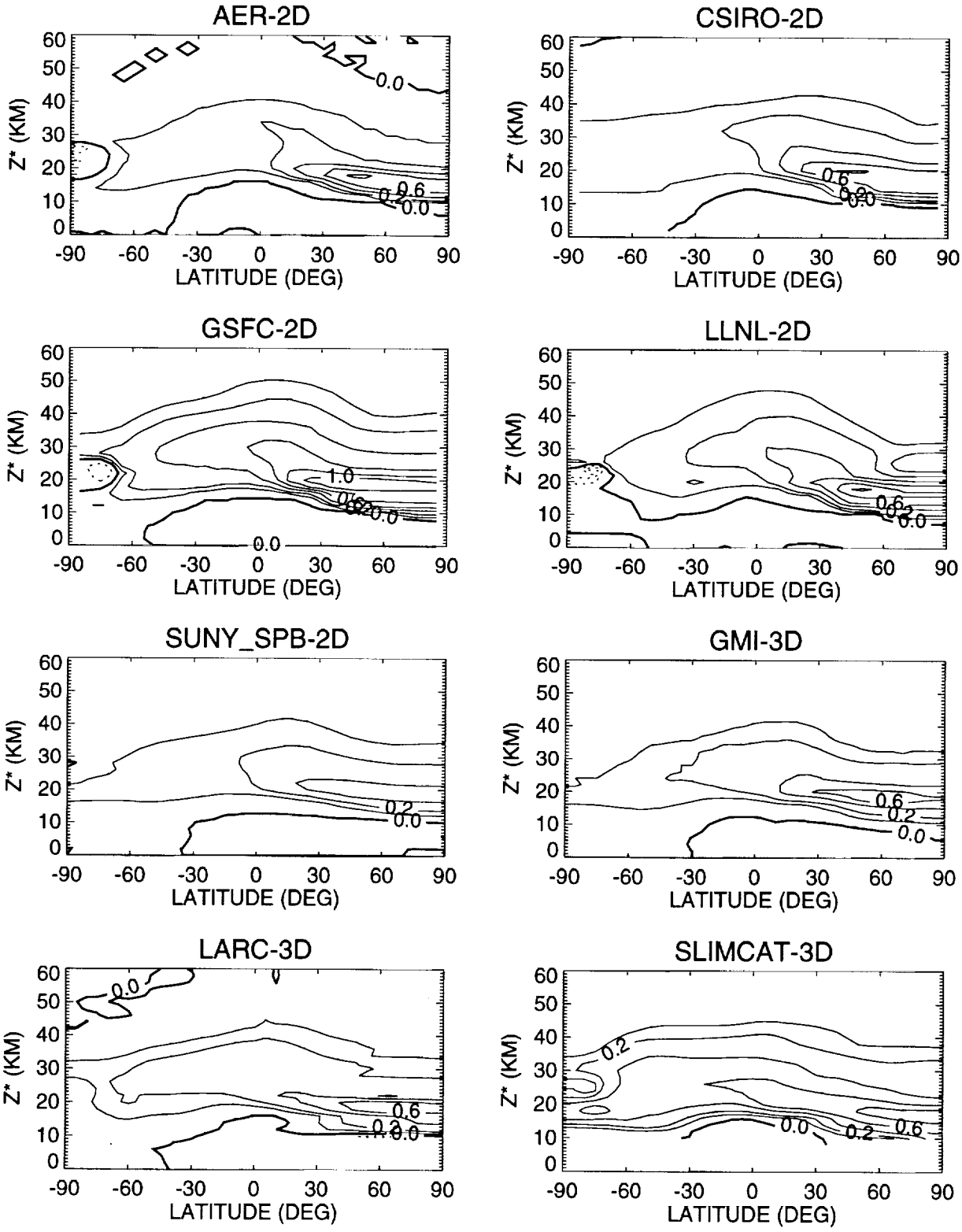
AER-2D
CSIRO-2D
GSFC-2D
LLNL-2D
SUNY_SPB-2D
GMI-3D
LARC-3D
SLIMCAT-3D
Z* (KM)
LATITUDE (DEG)
0
10
20
30
40
50
60
-90
-60
-30
0
30
60
90
0.0
0.2
0.6
1.0

Fig. 2

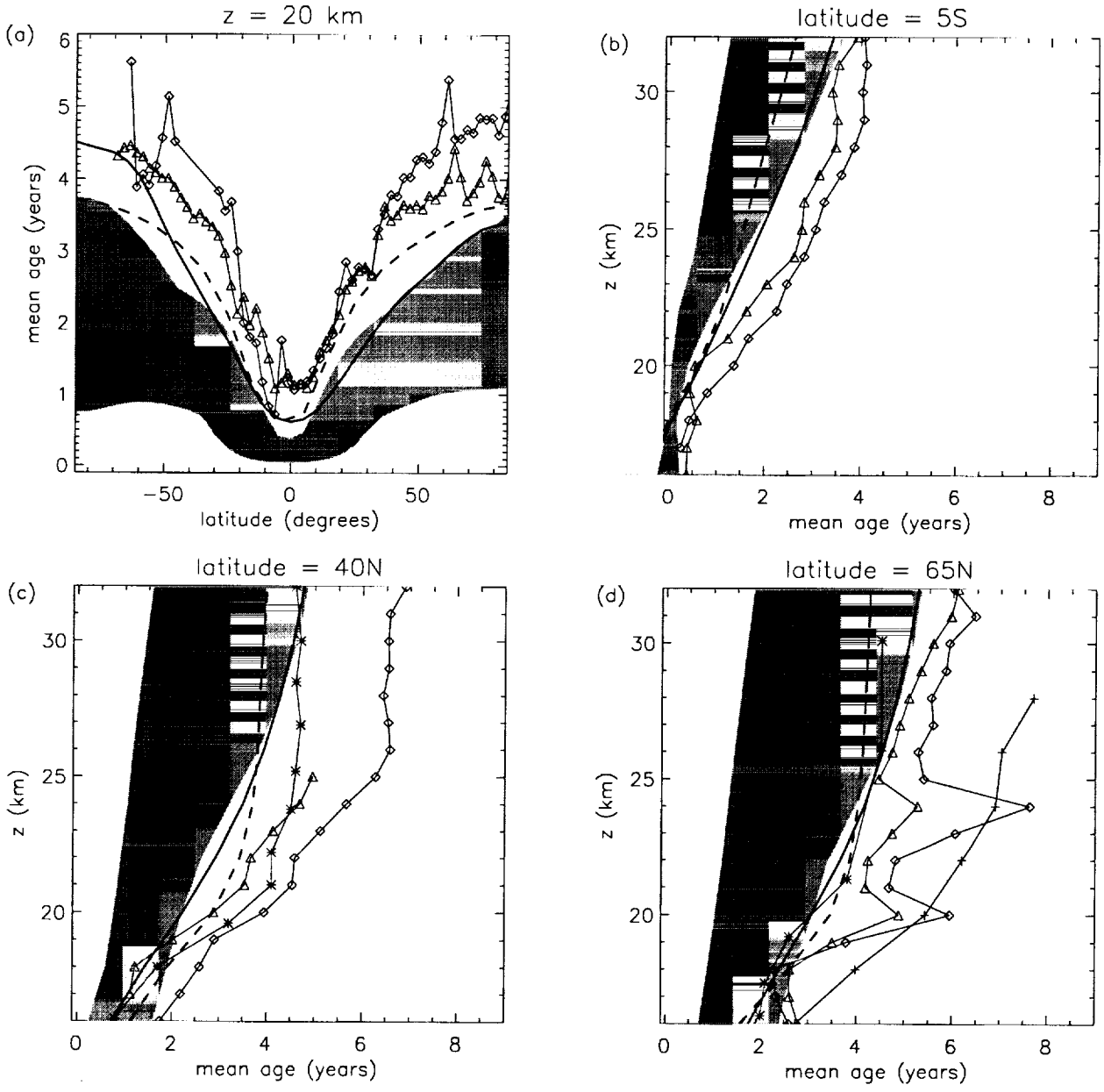
(a)
z = 20 km
mean age (years)
latitude (degrees)
(b)
latitude = 5S
z (km)
mean age (years)
(c)
latitude = 40N
z (km)
mean age (years)
(d)
latitude = 65N
z (km)
mean age (years)

Fig. 3

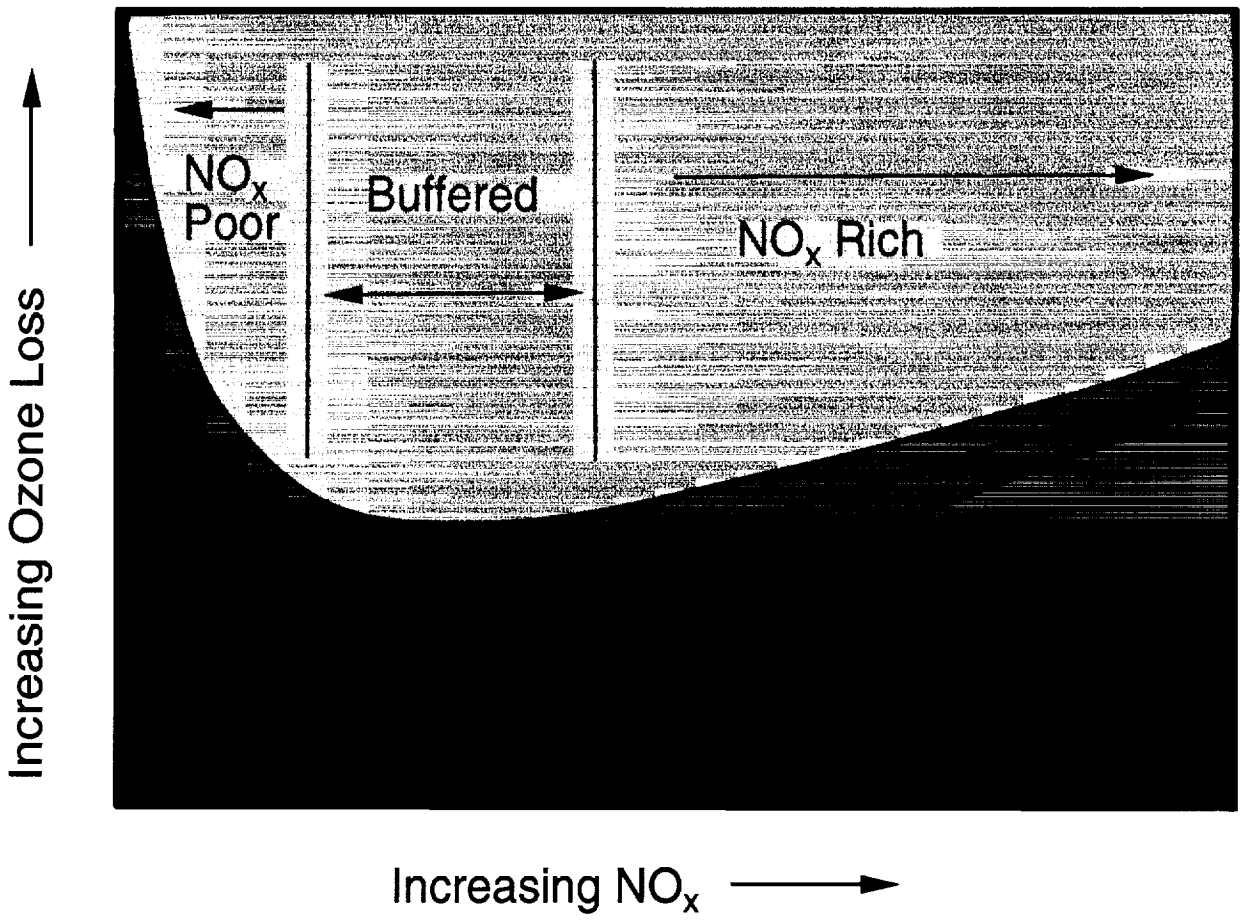
Increasing Ozone Loss
NOx Poor
Buffered
NOx Rich
Increasing NOx

Fig. 4