

Atmospheric impact of NO_x emissions by subsonic aircraft: A three-dimensional model study

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Abstract. Three-dimensional model calculations suggest that the world's fleet of subsonic aircraft has enhanced the abundance of nitrogen oxides in the upper troposphere by up to 20–35% and has produced a significant increase in the ozone concentration in this region of the atmosphere (4% in summer and 1% in winter). In year 2050, on the basis of current scenarios for growth in aviation, the concentration of NO_x at 10 km could increase by 30–60% at midlatitudes, and the concentration of ozone could be enhanced by 7% and 2% in summer and winter, respectively (relative to a situation without aircraft effects). The perturbation is not limited to the flight corridors but affects the entire northern hemisphere. The magnitude (and even the sign) of the ozone change depends on the level of background atmospheric NO_x and hence on NO_x sources (lightning, intrusion from the stratosphere, and convective transport from the polluted boundary layer) and sinks which are poorly quantified in this region of the atmosphere. On the basis of our model estimates, 20% of the NO_x found at 10 km (midlatitudes) is produced by aircraft engines, 25% originates from the surface (combustion and soils), and approximately 50% is produced by lightning. For a lightning source enhanced in the model by a factor of 2, the increase in NO_x and ozone at 10 km due to aircraft emissions, is reduced by a factor of 2. The magnitude of aircraft perturbations in NO_x is considerably smaller than the uncertainties in other NO_x sources.

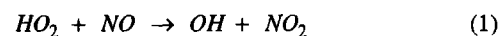
1. Introduction

Engines of subsonic aircraft are releasing into the upper troposphere and lower stratosphere significant quantities of chemical compounds. Nitrogen oxides (NO and NO₂, represented as NO_x) are of particular importance, since they have the potential to modify the ozone concentration near the tropopause and hence to perturb the radiative forcing on the climate system.

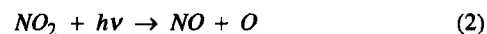
The amount of nitrogen oxides emitted by the 1990 fleet of subsonic aircraft and its spatial distribution in the global atmosphere was reported by NASA [Albritton *et al.*, 1993], based on a detailed estimation by Boeing and McDonnell Douglas. Corresponding estimates are also provided for fleets of subsonic and supersonic transports expected to fly in year 2015. As indicated in Table 1, the global injection of nitrogen into the atmosphere (released mainly between 10- and 12-km altitude) is estimated to represent 0.44 Tg(N)/yr and 0.82 Tg(N)/yr in 1990 and 2015, respectively. The overall NO_x emission index expressed in g(NO₂)/kg(fuel) is therefore 10.9 in 1990 and is reduced to 8.9 in the 2015 scenario, as engine technology is expected to improve. A recent estimate by the European Civil Aviation Conference (ECAC) [see *World Meteorological Organization*, 1995] suggests a current global aircraft emission of 0.85 Tg(N)/yr with an average NO_x emission index of 16.8.

In the stratosphere, nitrogen oxides destroy ozone directly through a catalytic cycle [Crutzen, 1970; Johnston, 1971] and

interact with other chemical families (e.g., hydrogen and chlorine) which also affect the ozone budget. In the troposphere and lower stratosphere, the oxidation of hydrocarbons in the presence of nitrogen oxides leads to the production of ozone [Crutzen, 1974; Chameides and Walker, 1976; Fishman *et al.*, 1979; Liu *et al.*, 1980; Logan *et al.*, 1981]. The key chemical cycle involves the conversion of nitric oxide (NO) into nitrogen dioxide (NO₂) by peroxy radicals (HO₂)



followed by the photolysis of nitrogen dioxide



and the recombination of atomic oxygen (O) with molecular oxygen (O₂). Reaction (1) is the rate limiting factor of this cycle. A reaction similar to (1) but where HO₂ is replaced by organic peroxy radicals (RO₂, where R is, for example, CH₃, C₂H₅, etc.) also leads to the production of ozone.

The impact of subsonic aircraft on tropospheric ozone is difficult to assess owing to large uncertainties in our understanding of chemical and dynamical processes in the upper troposphere and lower stratosphere. Poorly quantified processes include the natural production of NO_x by lightning, the vertical transport of trace constituents within convective clouds, stratosphere/troposphere exchanges, chemical reactions in clouds, heterogeneous removal processes, etc. In particular, the uncertainty associated with the convective parameterization significantly affects the estimated contribution of surface NO_x emissions to the upper tropospheric nitrogen budget. Although these uncertainties severely limit our ability to produce reliable assessments, numerical models can nevertheless be used to provide “best estimates” of aircraft perturbations, perform

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Table 1. Emission of Chemical Compounds by Existing (1990) and Projected (2015) Fleets of Subsonic Aircraft

	Year 1990 ^a	Year 2015 ^b
Fuel burn	134	304
NO _x emission ^c	1.46	2.70
Hydrocarbon emission ^d	0.352	0.338
CO emission	1.13	1.77

Units are teragrams per year.

^aIncludes the contributions of airliners and cargoes, turboprops, charters and military traffic on the basis of schedules provided by the May 1990 Official Airline Guide. Estimates of nonscheduled traffic are included.

^bFlight frequencies obtained by averaging regional growth rates predicted by Boeing and McDonnell Douglas.

^cexpressed as NO₂

^dexpressed as CH₄

sensitivity analyses, and underline the key factors which need to be measured or better formulated in future (and more advanced) models. The purpose of this brief report is to address several of these issues and to examine the effects of current and projected subsonic aircraft within the context of uncertainties in the nitrogen oxide budget.

2. Model Description

The potential impact of NO_x emissions by current and future fleets of subsonic aircraft on tropospheric ozone is assessed by using the IMAGES (Intermediate Model for Annual and Global Evolution of Species) model. This three-dimensional chemical transport model, described in detail by Müller and Brasseur [1995], simulates the global distribution of approximately 50 chemical species (including 6 nonmethane hydrocarbons and several related oxygenated organic species, including peroxyacetyl nitrate (PAN) and other organic nitrates) between the surface and a pressure level of 50 mbar (approximately 20 km). It accounts for surface emissions of biogenic and anthropogenic gases, for advective and convective transport for, subgrid mixing processes, and for chemical conversions (125 chemical and photochemical reactions), as well as dry and wet deposition processes. Convective transport is parameterized [Costen *et al.*, 1988] using the distribution of convective clouds provided by the International Satellite Cloud Climatology Project (ISCCP) [Rossow *et al.*, 1987; Rossow and Schiffer, 1991]. The resolution of the numerical grid is 5° in longitude and latitude and is variable in altitude: 25 levels in σ coordinates are defined with a vertical resolution of typically 50–100 m in the planetary boundary layer and 0.5–1 km in the free troposphere; a few levels are representative of the lower stratosphere. The σ coordinates are defined in IMAGES as

$$\sigma = \frac{p - 50 \text{ mbar}}{p_s - 50 \text{ mbar}}$$

where p and p_s are the pressure (in millibars) at a given level and the pressure at the surface, respectively. Meteorological and climatological parameters (temperature, winds, specific humidity, cloud distribution, lightning statistics, precipitations, ice sheet extent, ecosystem distribution, etc.) are taken from existing data analyses (e.g., European Centre for Medium-Range Weather

Forecasts (ECMWF)), satellite observations, and other compilations (see Müller and Brasseur [1995] for further details). Surface emissions were established for each month of the year on the basis of ecological and economic data and include technological contributions (fossil fuel burning, waste disposal, industrial activity, etc.), biomass burning, release by soils and foliage, exchanges with the ocean, etc. [Müller, 1992]. Note that in order to account for the intrusion of stratospheric ozone into the troposphere in spite of the fact that the model extends only up to 50 mbar, the ozone concentration is specified in the three uppermost levels of the model on the basis of ozonesonde measurements [Komhyr *et al.*, 1992; Logan and Kirchhoff, 1986].

The NO_x sources represented in this model (see Table 2) include the contributions of (1) fossil fuel combustion, (2) biomass burning, (3) soil emissions, and (4) lightning associated with thunderstorms. The production of NO following nitrogen fixation by lightning flashes requires special attention because it injects substantial quantities of NO_x in the layers where aircraft are cruising. This source is poorly quantified, with most estimates ranging from approximately 1 Tg(N)/yr to 20 Tg(N)/yr [Tuck, 1976; Chameides *et al.*, 1977; Peyroux and Lapeyre, 1982; Logan, 1983; Borucki and Chameides, 1984; Liaw *et al.*, 1990]. In this study, we adopted a global lightning source of 5 Tg(N)/yr, which is distributed geographically and seasonally according to lightning flashes detected by satellites [Turman and Edgar, 1982]. We assume that the number of NO molecules produced per unit time and volume is constant with altitude inside convective clouds. The model accounts for heterogeneous conversion of N₂O₅ into HNO₃ on the surface of sulfate aerosols [Ehhalt and Drummond, 1982; Dentener and Cruzen, 1993].

Most of the results presented in this paper will be shown as zonal means or, when displayed as a function of latitude and longitude, on the surface corresponding to $\sigma = 0.2$ (approximately 240 mbar, or 10-km altitude) where the chemical impact of aircraft emissions appears to be the largest. At this level, the mixing ratio of ozone derived from IMAGES is typically of the order of 80–100 parts per billion by volume (ppbv) in the northern hemisphere. The mixing ratio of NO_x (Plate 1) varies

Table 2. Global Budget of Atmospheric Nitrogen Compounds in the IMAGES Model

	Value
Sources	
Surface emissions (anthropogenic, biomass burning, soils)	35.0
Lightning	5.0
Aircraft (troposphere)	0.4
Oxidation of nitrous oxide (stratosphere)	0.2
Total	40.6
Sinks	
HNO ₃ wet deposition	17.7
HNO ₃ dry deposition	9.7
Organic nitrate deposition	8.3
NO _x dry deposition	4.1
Peroxyacetyl nitrate dry deposition	0.4
HNO ₄ wet deposition	0.4
Total	40.6
Burden, teragrams of nitrogen	2
Residence Time, days	18

Unless otherwise indicated, values are teragrams of nitrogen per year.

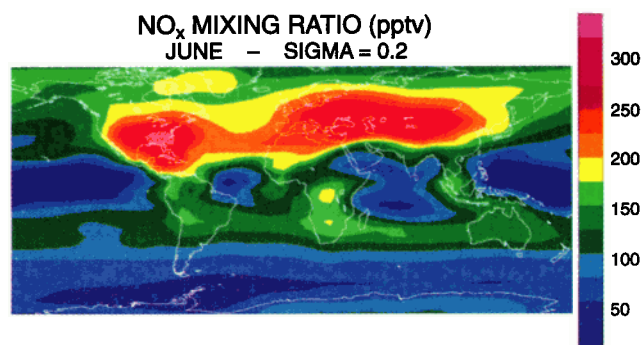


Plate 1. Mixing ratio of NO_x ($\text{NO} + \text{NO}_2$) calculated for June conditions on the level corresponding to $\sigma = 0.2$ (approximately 240 mbar, or 10-km altitude).

from 40 parts per trillion by volume (pptv) in the tropics to 325 pptv over the southern United States, with values close to 250 pptv over Europe and 180 pptv in the North Atlantic corridor. *Drummond et al.* [1988] observed NO mixing ratios of 120–150 pptv in the vicinity of the tropopause (corresponding approximately to 160–200 ppbv of NO_x). In October 1993, *Schlager et al.* [1994] observed in the upper troposphere NO mixing ratios of 80–180 pptv east of Scotland and of 200–500 pptv within the North Atlantic flight corridor. *Ridley et al.* [1994] reported NO_x mixing ratios of 120–500 pptv in the upper troposphere over New Mexico during summer. Observations made by *Weinheimer et al.* [1994] during the Airborne Arctic Stratospheric Experiment suggest NO_x ($\text{NO} + \text{NO}_2$) mixing ratios near the tropopause varying between approximately 30 pptv over the pole to 90 pptv at 40°N , with values reaching occasionally 140 pptv in the vicinity of frontal systems.

3. Modeled Impact of Subsonic Aircraft

The relative change in the concentration of NO_x resulting from the presence of subsonic aircraft (1990 fleet) is presented in Plates 2a and 2b for July conditions. The largest perturbation shown in the zonal mean representation (25%) is located near the 10-km level between 50° and 60°N latitude, with small effects above 15 km and below 5 km as well as in the entire southern hemisphere. As shown by Plate 2b, the largest impact (35%) is found over northern Europe, including a part of the North Atlantic Ocean. A secondary maximum (25%) is found over the United States. *Beck et al.* [1992] estimate that in April, the NO_x injected by aircraft, in the absence of the lightning source, contributes to 30–50% of the NO_x present at 8–12-km altitude and 30°N – 60°N . *Ehhalt et al.* [1992] suggested that on the average, 30% of upper tropospheric NO_x is provided by aircraft and that NO_x lofted from the planetary boundary layer is another major source, while *Kasibhatla* [1993] derived from his three-dimensional model study that it is not necessary to invoke fast convective transport to explain the observed NO_x mixing ratios near the tropopause. In the IMAGES model, the origin of midlatitude NO_x at 10 km is the following: 15–20% from aircraft emissions, 30–60% from lightning, and approximately 25% from surface sources (20–22% from man-made combustion, 2–4% from soils, and 2–4% from biomass burning). At 5 km (30° – 60°N), 5–30% of the NO_x is provided by lightning sources, 75–85% by surface emissions (70–80% by man-made combustion), and approximately 5% by aircraft operations (see J.-F. Lamarque

et al., manuscript in preparation, 1995). The range in these estimates represents the magnitude of seasonal variations.

The only significant impact of subsonic aircraft on the ozone abundance appears to be limited to the northern hemisphere (Plates 3a and 3b) and is most pronounced in summer when

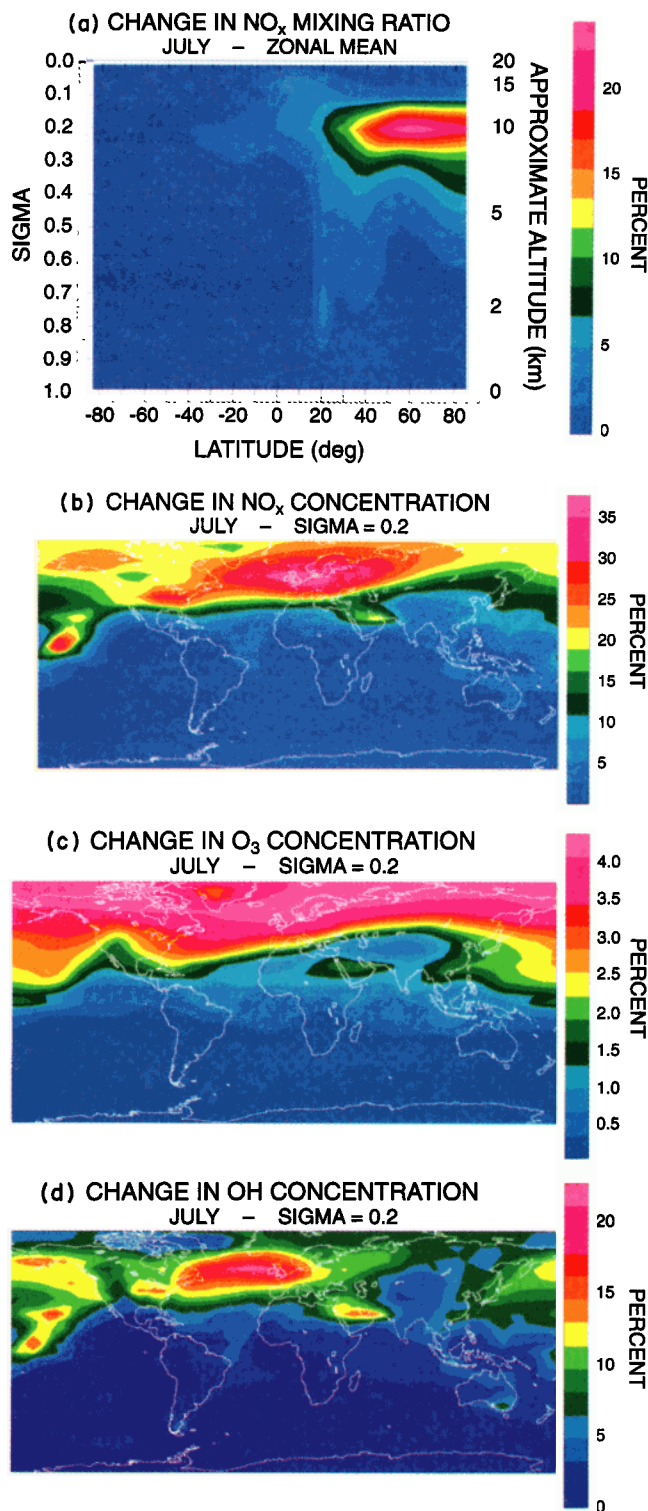


Plate 2. Relative change (in percent) in (a) the zonally averaged NO_x concentration calculated as a function of latitude and height (July) for the 1990 fleet of subsonic aircraft; (b) the NO_x concentration at $\sigma = 0.2$ level; (c) the ozone concentration at the same level; and (d) the OH concentration at the same level.

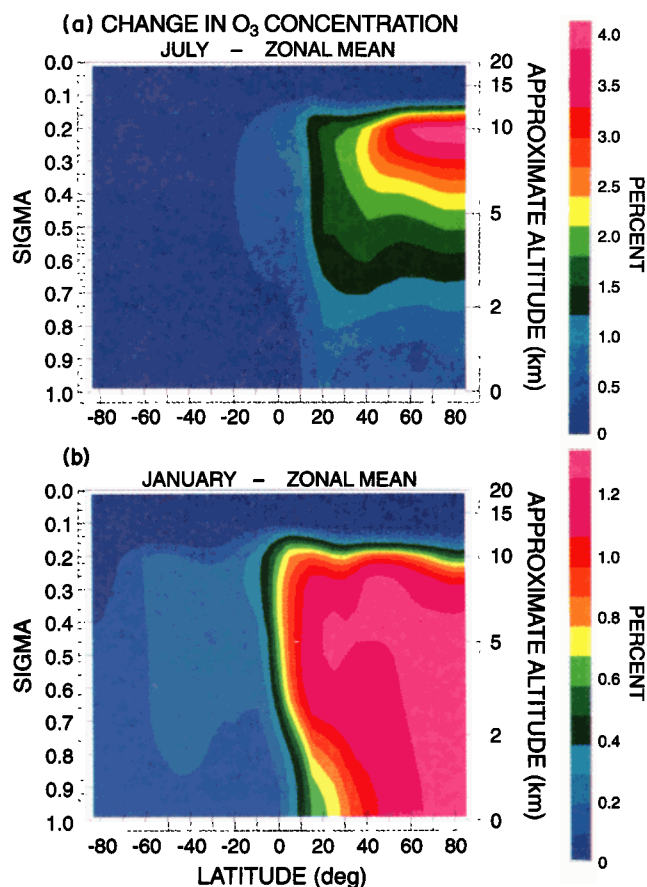


Plate 3. Calculated change in the zonally averaged ozone concentration as a function of latitude and height for the 1990 fleet of subsonic aircraft for (a) July and (b) January.

reactions (1)–(2) are most efficient. The zonally averaged ozone change reaches a maximum of 4% in July (10 km, north of 50°N) and 1.3% in January. Note that the perturbation effect on ozone propagates to lower altitudes in winter, especially at high latitudes, where the photochemistry is less effective and the downward transport efficient. On the $\sigma = 0.2$ level, the maximum ozone change in July (4.5%, see Plate 2c) is found over northern Canada, northern Europe, and northern Siberia. However, the ozone perturbation is spread rather uniformly over the entire northern hemisphere, rather than being concentrated near the most active flight corridors. The change in upper tropospheric OH (Plate 2d) is largest over the North Atlantic corridor, where it reaches approximately 20% during summer. The effect of NO_x emissions on other global parameters (photochemical lifetime of methane, photochemical production of ozone, and wet deposition of nitric acid) is given in Table 3.

The impact of NO_x released by aircraft varies in a nonlinear way with the background level of NO_x in the atmosphere. Figure 1 shows the calculated net production of ozone at approximately 10 km as a function of the NO_x abundance. In curve A, which corresponds to the assumptions made in the model of *Ehhalt et al.* [1992], the adopted mixing ratios are 84 ppbv for ozone, 73 ppbv for carbon monoxide, 1.6 ppmv for methane, and 50 ppmv for water vapor. In addition, the effect of nonmethane hydrocarbons is neglected, and the solar diurnal cycle is ignored. The calculated production of ozone is substantially modified when the mixing ratio of water vapor is doubled (100 ppmv; curve B), when, in addition, the effects of hydrocarbons are taken into account (curve C), and finally, when the diurnal cycle is included in the model calculation (curve D). Since the maximum ozone production in the four curves corresponds to NO_x concentrations close to background atmospheric values but differ significantly with the assumptions made (150 pptv for curve A, 170 pptv for curve B, 190 pptv for curve C, and 270 pptv for curve D), the ozone response to aircraft NO_x varies significantly with model assumptions. The conditions adopted in the IMAGES model correspond to curve D. With the chemical scheme used in the IMAGES model, the net ozone production rate at 10 km reaches a maximum for a NO_x mixing ratio of approximately 280 pptv. As the mixing ratios predicted by the model in the upper troposphere are usually lower than this value, the additional NO_x provided by aircraft leads to an enhanced net ozone production and hence to an increased ozone concentration. For background mixing ratios of NO_x larger than approximately 270 pptv, the ozone density would be reduced as a result of NO_x injections. In the model of *Ehhalt and Rohrer* [1994], the corresponding transition should occur for a NO_x mixing ratio of approximately 130 pptv.

The change in the radiative forcing in the northern hemisphere resulting from the calculated perturbation induced by the 1990 fleet of subsonic aircraft is, on the average, of the order of 0.02 W/m^2 in summer and 0.01 W/m^2 in winter, with a local maximum of approximately 0.04 W/m^2 at midlatitudes in June and July. This climatic perturbation is significantly smaller than the total radiative forcing which has been produced by other greenhouse gases since the preindustrial era. The slight decrease in the methane abundance produced by the release of NO_x by aircraft does not modify significantly the radiative forcing. A reduction of 1% in the atmospheric abundance of methane (corresponding at steady state to a 1% decrease in the CH_4 lifetime; see Table 3) would offset the warming caused by enhanced ozone concentrations by only 0.001 W/m^2 .

In order to determine the significance of the aircraft emissions on the chemical composition of the atmosphere, the magnitude of the chemical perturbations caused by aircraft emissions was compared with the impact of potential changes (uncertainties) in other NO_x sources. The model shows, for example, that if the anthropogenic emissions of NO_x at the surface are uniformly increased by 25%, the mixing ratios of NO_x at midlatitudes in the

Table 3. Annually and Globally Averaged Differences of Perturbations Caused by NO_x Injections

	1990 Aircraft Fleet	2015 Aircraft Fleet	Doubling of Lightning	Anthropogenic Surface Sources Increased by 25%
CH_4 chemical lifetime	-1.0	-1.9	-15	-3.0
O_3 photochemical production	+0.7	+1.4	+12	+3.4
HNO_3 wet deposition	+2.0	+3.6	+28	+15

Values are in percent.

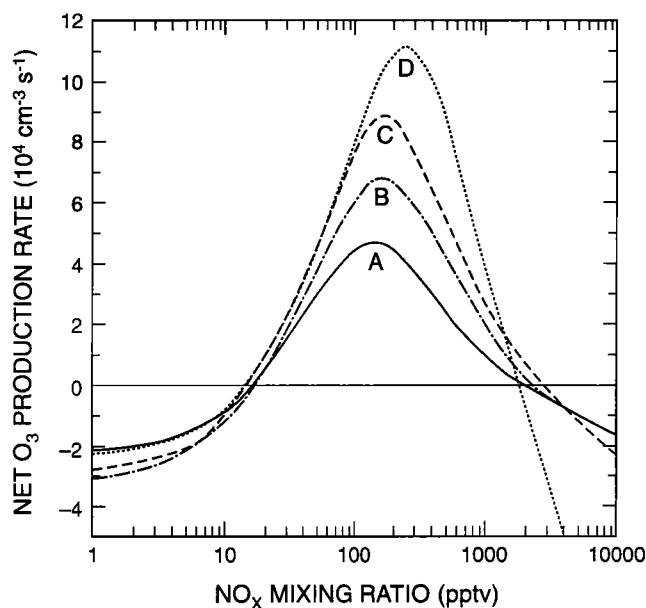


Figure 1. Net ozone production rate in cubic centimeters per second calculated as a function of the atmospheric NO_x mixing ratio (in parts per billion by volume) at 50°N at 10-km altitude in July for clear-sky conditions. For detailed explanations, see text.

northern hemisphere (zonal mean summer values) increase by approximately 10% between 4- and 6-km altitude where NO_x has been transported by convection. At 10 km, the calculated change is only 3–5%, compared to the aircraft effect of approximately 25% in the northern hemisphere. If on the other hand, the NO_x source by lightning is doubled, the abundance of NO_x is substantially enhanced in the upper troposphere and lower stratosphere (Plate 4a) and so is the concentration of ozone (Plate 4b), particularly in the tropics (18% at the equator, 6% at middle and high latitudes at 10 km in July). In the most active flight corridors of the northern hemisphere, the NO_x concentration increases by typically 40–50% owing to the doubling in the lightning source; this value depends, however, on the assumption made for the vertical distribution of the lightning source, as well as on its magnitude. Thus the uncertainty associated with this source of nitrogen oxides in the troposphere has major consequences for several chemical processes and for the assessment of the aircraft impact. If the model predictions for aircraft perturbations presented above are repeated with a doubled strength of the lightning source (10 $\text{Tg}(\text{N})/\text{yr}$), the maximum increases in the zonally averaged NO_x and ozone perturbations in July are reduced to 15% and 2.4%, respectively. It is clear from Table 3 that the impact of aircraft is considerably smaller than the uncertainties in the other NO_x sources, especially the source associated with lightning. The chemical perturbation per unit mass of NO released is, however, larger in the case of aircraft injection than for surface emissions. For example, the photochemical lifetime of methane (which is a measure of the oxidizing capacity of the atmosphere) changes by 2.5%/Tg(N) released by aircraft and by only 0.6%/Tg(N) released at the surface.

Finally, when adopting for the NO_x emissions a scenario intended to represent the situation in year 2015 [Albritton *et al.*, 1993], the concentration of NO_x increases by 35–60% near 10 km at midlatitudes, compared to a model simulation in which the

presence of aircraft is ignored. The corresponding change in the ozone concentration at 10 km (assuming a constant lightning source of 5 $\text{Tg}(\text{N})/\text{yr}$) is predicted to be approximately 7% and 2% at northern midlatitudes in summer and winter, respectively. Thus over the next 20 years, the ozone concentration in the midlatitude upper troposphere could further increase by approximately three quarters of the increase calculated for the period over which aviation has developed.

4. Conclusions

Calculations made with the three-dimensional IMAGES model suggest that the abundance of NO_x in the upper troposphere has increased significantly in the northern hemisphere as a result of subsonic aircraft operations. The magnitude of this increase depends on the adopted aircraft emissions, as well as the estimated natural sources of NO_x which remain poorly quantified (in particular the production of NO by lightning flashes in thunderstorms). The increase in the zonally averaged NO_x concentration due to the 1990 aircraft fleet is estimated by the model to be as high as 25% at midlatitudes in the northern hemisphere. It could, however, be a factor of 2 higher if the emission inventory reported by ECAC was adopted (rather than that of Albritton *et al.* [1993]) or a factor of 2 lower if the specified lightning source was 10 $\text{Tg}(\text{N})/\text{yr}$ rather than 5 $\text{Tg}(\text{N})/\text{yr}$. With the scenario adopted in IMAGES, the calculated maximum increase in the ozone abundance is 4% in summer and

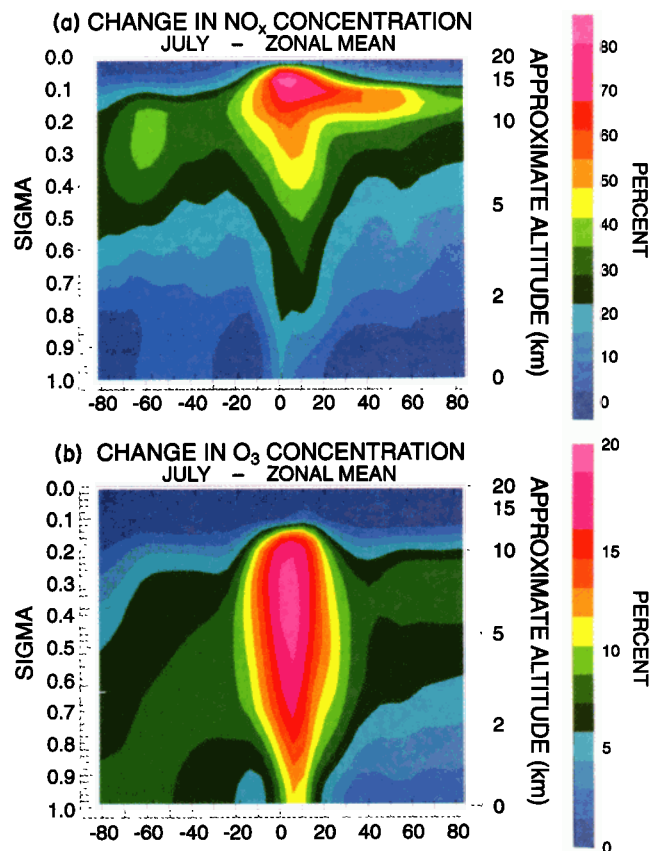


Plate 4. (a) Relative change (in percent) in the zonal averaged NO_x concentration (July) for a spatially uniform doubling in the NO_x source by lightning flashes and (b) corresponding increase in the ozone concentration.

1.3% in winter. Finally, the radiative forcing produced by the perturbation in ozone appears to be limited. The radiative impact of soot particles, sulfate aerosols, and water contrails produced by the aircraft could potentially play a more significant role. In order to reduce the uncertainties in the current estimates, observational programs will have to provide surveys of background NO_x in the upper troposphere and to better quantify natural sources and sinks of NO_x , in particular the production of NO by lightning.

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