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Effects of lightning and convection on changes in tropospheric ozone

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Abstract

A global 3-dimensional chemical tracer model (CTM) has been used to study the impact on tropospheric ozone caused by NOx emissions from today's fleet of subsonic aircraft (0.52 Tg(N)/yr), due to uncertainties in sources of upper tropospheric NOx from lightning and deep convection.

Three sets of two CTM experiments have been performed, with and without emissions from aircraft. A reference set with normal convection and 12 Tg(N)/yr from lightning, a set with reduced lightning source (5 Tg(N)/yr), and one set with reduced convective activity. A zonally homogeneous increase in upper tropospheric ozone north of 40°N, reaching 5-6 ppbv during May was found in the reference case.

Reduced emissions from lighting lead to 50-70 % higher enhancement of ozone at northern mid-latitudes during summer, due to lower background concentrations of NOx and more efficient ozone production. Reduced convective mixing lead to a 40% increased enhancement in aircraft induced ozone at northern mid-latitudes and 150% in the tropics. In this case background NOx levels were higher in the upper troposphere, but the decreased ozone production efficiency was compensated by decreased downward mixing of ozone produced by emissions from aircraft.

Acknowledgements

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1 Introduction

Emissions of NOx from subsonic aircraft have the potential to increase the ozone in the upper troposphere significantly. However, there are substantial uncertainties in the NOx budgets in this region of the atmosphere. In this study we investigate how uncertainties in to of the major NOx sources, namely lightning and convection influence the impact of aircraft.

Changes in ozone in the upper troposphere will have significantly larger effects on radiative forcing of climate than changes in other altitude regions of the atmosphere (Wang and Sze, 1980; Lacis et al., 1990; IPCC 1992).

Model studies of the impact of increased emissions of ozone precursors since pre-industrial times indicate that the increase in tropospheric ozone has caused a global averaged radiative forcing of 0.2-0.6 W/m^2 (IPCC-95) or about 20% of the increase in forcing caused by the increase in the concentrations of well-mixed greenhouse gases (Hauglustaine et al., 1994; Lelieveld and van Dorland 1995; Muller and Brasseur 1995; Forster et al., 1996; Berntsen et al., 1997).

Ozone in the troposphere is formed in situ by chemical reactions involving nitrogen oxides (NOx = NO + NO_2), methane (CH_4), carbon monoxide (CO), non-methane hydrocarbons (NMHCs) and sunlight (Crutzen, 1973), and by intrusion of ozone rich air from the stratosphere. The efficiency of the ozone formation per NOx molecule oxidised is a highly non-linear function of the ambient NOx levels (e.g. Isaksen et al., 1978; Lin at al., 1988). Emissions of NOx from aircraft in the upper troposphere takes place in an environment with generally lower levels of NOx than in the continental planetary boundary layer (PBL). Due to the combination of chemical and radiative effects NOx from aircraft is expected to have a much higher impact on climate than similar emissions from surface sources (e.g. WMO 1995; Fuglestvedt et al., 1996).

NOx emissions generally increase OH concentration and thereby decrease methane concentrations. The radiative forcing of the lower methane concentrations tends to be of the opposite sign and of similar magnitude as the impact of ozone for *surface* sources of NOx. For NOx from aircraft, however, the effect on methane is smaller due to lower humidity and temperatures in the upper troposphere (Fuglestvedt et al., 1996).

The total source of NOx from aircraft for the present fleet of subsonic aircraft has been estimated to be of about 0.5 Tg(N)/yr, with a projected increase in fuel use of 3-5%/yr until 2015 (Jenkins, 1996). Other sources of NOx in the upper troposphere is lightning (2-20 Tg(N)/yr), transport from the PBL by deep convection, input from the stratosphere (<1 Tg(N)/yr) and recycling of NOy species (PAN, HNO_3, HO_2NO_2) through thermal decomposition, photolysis and possibly heterogeneous processes (Hauglustaine et al., 1996).

Several model studies have addressed the question of the impact of the emissions from aircraft on upper tropospheric NOx, NOy, and ozone (Brasseur et al., 1996; Schumann, 1997a; Schumann, 1997b; Wauben et al., 1997; Stevenson et al., 1997, Dameris et al., 1998). However, due to the non-linear dependence of the ozone formation, uncertainties in the other sources of upper tropospheric NOx has a significant influence on the estimated changes in ozone distribution.

In this work we use the University of Oslo 3-D global chemical tracer model (CTM) (Berntsen and Isaksen, 1997; Jaffe et al., 1997, Berntsen et al., 1997) to study how the uncertainties in deep convection and lightning sources of upper tropospheric NOx influence the estimated increase in upper tropospheric ozone from aircraft.

The impact of NOx emissions from aircraft on upper tropospheric ozone in a base case with 12 Tg(N)/yr from lightning and with standard convective mixing (from the NASA/GISS GCM, see Prather et al., 1987 for details) have been compared with the corresponding impact of aircraft in two perturbation cases. In case *LL* the lightning source of NOx is reduced to 5 Tg(N)/y, while in case *LC* the convective activity in the model is reduced by 67%.

2 Model Description

The model has been discussed in detail elsewhere (Prather et al., 1987; Berntsen and Isaksen, 1997; Berntsen et al., 1997), hence only a short description will be given here.

The CTM has a resolution of 8° by 10° (latitude vs. longitude) and 9 vertical layers below 10 hPa. The vertical resolution in the vicinity of the tropopause is about 3 km at mid and high latitudes, and about 4.5 km in the tropics. The transport formulation includes three-dimensional advection, calculated by the very accurate "second order moments" method of Prather (1986), as well as rapid vertical mixing by convective transport (Prather et al., 1987). The strength of the deep convective transport is quite uncertain. Jacob et al. (1997) have compared vertical mixing of radon (^{222}Rn) in various CTMs, including the GISS/H/I model which has a very similar transport parameterisation as our CTM. They conclude that the simulated deep convection in the troposphere is within the constraints offered by seasonal averages of ^{222}Rn concentrations at different altitudes.

A comprehensive photochemical scheme with 55 chemical components and 120 gas-phase reactions has been incorporated to simulate the photochemistry of the troposphere. For this work the chemistry scheme has been extended to include degradation of propane, which gives acetone which has been found to be a major constituent in the upper troposphere (Singh et al., 1995; Arnold et al, 1997). Within the troposphere, the chemistry with a full diurnal cycle is calculated with a quasi steady-state approximation method (QSSA), with iterations for the short-lived species (Hesstvedt et al., 1978).

In the stratosphere, concentrations of ozone, NOx and HNO_3 are fixed according to the observed distribution of these species. The circulation within the whole model domain up to 10 hPa then generates an internally consistent cross-tropopause flux. The photolysis rates have been calculated with the two-stream algorithm of Kylling et al. (1995). The model uses pre-calculated meteorological input data with 8-hour time resolution from a one year climate simulation with the NASA/GISS GCM. The continuity equation is solved by an operator splitting procedure with a one-hour timestep for the transport, and 30 minutes for the chemistry.

In this study chemistry calculations have been done up to about 18.5 km altitude (σ = 0.061) in the tropics (equator-ward of 32°), and up to about 13.7 km (σ = 0.144) at higher latitudes. Above these altitudes the concentrations of O_3, NOx and HNO_3 was prescribed as described in Berntsen et al. (1997).

2.1 Convection

Rapid vertical transport within deep convective cells can transport air originating in the PBL to the upper troposphere within less than an hour. Short-lived species like NOx can therefore be transported to the upper troposphere in significant amounts. Strand and Hov (1996) estimate in a 2-D zonally averaged model that rapid mixing within convective cells and frontal zones contribute about 0.7Tg(N)/yr of NOx from the PBL to the free troposphere for the month of July. Köhler et al. (1997) find in a 3-D CTM a contribution from surface sources contribute to the upper tropospheric NOx burden at northern mid-latitudes of about 10 and 25% for January and July respectively.

An important aspect of convective transport of NOx from the surface is the positive correlation between regions of intense deep convection and elevated concentrations of NOx in the PBL (i.e. continental regions) at least at northern mid-latitudes where most of the emissions from aircraft take place. Due to these factors and the non-linear ozone chemistry, we believe it is important to apply a 3-D CTM in this kind of study.

The parameterisation of vertical mixing due to convection in this CTM is described in Prather et al. (1987). It includes rapid updrafts in the thermals and slower subsidence of the surrounding air, but not downdrafts. In this CTM the net effect of convective mixing is to deposit about 120 Gg(N)/month to the upper troposphere of the northern hemisphere (390-150 hPa) for the month of July. Correspondingly, lightning (reference case, 12 Tg(N)/yr globally) and aircraft are found to contribute about 390 and 42 Gg(N)/month respectively.

A major source of the discrepancies between earlier model estimates of the impact of aircraft emissions is differences in the terminal height of upward transport in convective cells. In some of the models the emissions from aircraft probably occur at the same altitude as the convective cloud tops (i.e. in the same vertical model layer), while in other models the two sources are normally separated. Due to the non-linearity of the chemistry the impact will be largest in the latter case. To examine how this works in this CTM, we have calculated the monthly mean deep convective mass flux from below into each vertical layer. Figure 1 shows a comparison of the zonal mean distribution of the convective mass flux for July and the corresponding distribution of aircraft emissions.

The emissions from aircraft have a maximum in model layer 7 (≈10.3-13.7 km altitude) at 45°N, decreasing by about 75% to the model layer below (≈7.4-10.3 km altitude). At northern mid-latitudes during summer, the maximum convective flux is deposited in layer 6. However, there is a significant amount (35-40% of the value in layer 6) is deposited at the altitude of maximum aircraft emissions as well. In the tropics there both the convection and the aircraft emissions have the largest impact at the same altitudes. The maximum in convective flux to the middle troposphere (4-6 km) found in the southern hemisphere, is associated with frontal activity in extra tropical cyclones. A similar maximum is found in the northern hemisphere during winter.

It is difficult to compare the distribution of convective mass fluxes given in figure 1 with observations. Still, a comparison with the frequency distribution of cloud top pressures from ISCCP (Rossow and Schiffer, 1991) shows that the ratio between the observed frequency of cloud top hights reaching 310 and 180 hPa respectively in July is very similar to the ratio between the convective mass fluxes to layer 6 and 7 in the CTM. This indicates that the altitude distribution of the vertical mixing by deep convection in the CTM is correct. The amount of transport of air from the PBL aloft can of course not be inferred from the ISCCP data. Thus, the comparison above does not provide a validation of the vertical mixing by deep convective clouds in the CTM.

2.2 Experimental setup

A total of six, 16 months model experiments have been performed for this study. Two experiments for reference case with and without emissions from aircraft (called case R_A and R_0 respectively), two experiments with a decreased lightning source (5 Tg(N)/yr instead of 12 Tg(N)/yr as in the reference case) (cases LL_A and LL_0), and two experiments with reduced convection (cases LC_A and LC_0). In the LC experiments the number of convective events (i.e. the convective mass flux) was reduced by 67%.

The rationale for choosing 5 and 12 Tg(N)/yr from lightning was that 5 Tg(N)/yr represents the most commonly used figure in previous studies, while 12 Tg(N)/yr is the result obtained from a recent and well documented study (Price et al., 1997a; Price et al., 1997b). To study the uncertainty caused by convective transport, we chose to reduce the activity by 67% based on results obtained by Jacob et al. (1997).

In a comparison of global atmospheric transport in 7 CTMs they found that the simulated concentration of ^{222}Rn in the upper tropospheric varied by approximately this amount between the models both at northern mid-latitudes and in the tropics. In the tropics the GISS/H/I model which has a transport which is very similar to our CTM gave the highest concentrations, while at northern mid-latitudes the GISS/H/I model gave results about the same as the mean of the 7 CTMs.

3 Emissions

3.1 Surface and aircraft emissions

The available emission inventories from the IGAC/GEIA database (for 1985 conditions) have been used in this work, and have been supplemented by other well-documented sources as given in Table 1.

Of particular importance for this study is the emission inventories for aircraft and lightning. The aircraft emissions are taken from the ANCAT/DLR-2 database (Nüsser and Schmitt, 1996), and include seasonal estimates of NOx emissions from civil and military air traffic (figure 1).

Table 1: Global annual emission rates applied in the model.

Species	Source	Annual emission rate
NOx (Tg(N)/yr)	Fossil Fuels[1]	21.0
	Biomass burning[2]	4.4
	Soils[1]	5.5
	Lightning[3]	12.2
	Aircraft[4]	0.52
	Total	46.4
CO (Tg/yr)	Anthropogenic[2]	893
	Biomass burning[2]	714
	Soils[2]	387
	Oceans[2]	378
	Total	2362
Isoprene[1] (Tg/yr)	Natural	570
Other NMHCs[5]	Natural	64.8
	Anthropogenic	114

[1] From the GEIA inventory (http://blueskies.sprl.umich.edu/geia)
[2] From Müller (1992) and Müller and Brasseur (1995)
[3] From Price et al. (1997a)
[4] From the ANCAT/DLR-2 database (Nüsser and Schmitt, 1996)
[5] See Berntsen and Isaksen (1997) and references therein.

3.2 Lightning

3.2.1 Total emissions

Emissions of NOx from lightning have been estimated in a number of studies, and the estimated total global source varies between 1 Tg(N)/yr (Levine et al., 1981) and 100 Tg(N)/yr (Franzbleu and Popp, 1989). Recent estimates by WMO (WMO, 1995) and Lee et al. (1997) give a range of 2-20 Tg(N)/yr, with 5-7 Tg(N)/yr as most likely. In previous CTM studies, a lightning source of 3-8 Tg(N)/yr is applied (Dentener and Crutzen, 1993; Müller and Brasseur, 1995; Lelieveld and van Dorland, 1995; Roelofs and Lelieveld, 1995; Strand and Hov, 1996; Wauben et al., 1997; Berntsen et al., 1997). Recently, Price and co-workers (Price et al., 1997a) have re-evaluated the NOx source from lightning and arrived at a significantly higher global annual emission rate of 12.2 Tg(N)/yr. Price et al. (1997b) arrive at a similar high estimate applying an independent method, using the global atmospheric circuit to put a constraint on the global lightning activity.

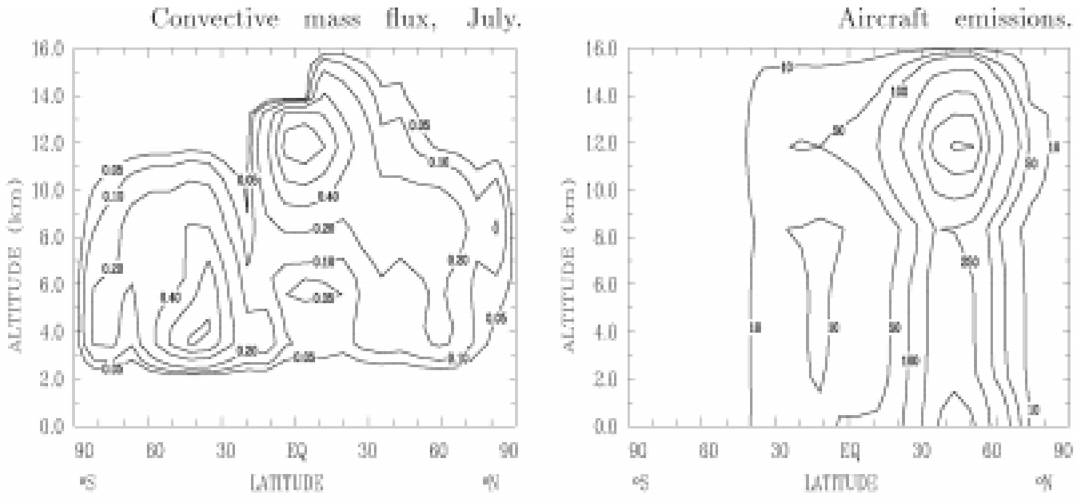

Figure 1. Left: Monthly mean zonal averaged deep convective mass flux (of air) from below to each model layer. Values are normalised, giving the gridcell with the highest flux a value of 1.0. Isolines at 0.05, 0.1, 0.2, 0.4, 0.6, 0.8 and 1.0. Right: Zonal mean distribution of aircraft emissions of NO. Isolines at 10, 50, 100, 250, 500, 750 and 1000 molecules s^{-1} cm^{-3}

Lightning emissions of NOx for this study has been included in the CTM by starting out with the monthly averaged source given on a 2.5° by 2.5° grid from Price et al. (1997a) and the input data on deep convection from the GISS GCM (Hansen et al., 1983). Emission of NOx from lightning takes place in connection with rapid vertical mixing in deep convective cells.

Based on the GCM data and a parameterisation of lightning activity as a function of cloud top height for continental and marine clouds respectively (Price and Rind, 1993) we calculate the longitudinal and temporal distribution of lightning. We then use this to distribute the zonally averaged emissions from Price et al. (1997a) as a function of longitude and time (8-hour resolution) according to the calculated lightning activity in each grid column. The details of the methods are given in the appendix. This procedure also assures that the lightning emissions are inserted into columns where strong vertical mixing by deep convection is occurring at the same time.

3.2.2 Vertical distribution

The rapid vertical mixing of NOx from lightning in thunderstorms has two consequences with respect to the impact of the emissions. First, the NOx from the lightning is redistributed vertically. Downdrafts within the storm might deposit a fair amount of NOx close to the ground, while updrafts transport the NOx into the top of the cloud, where it is detrained into the surrounding air. Also, the higher energy per stroke in CG strokes than in intra cloud strokes (Price et al. 1997a), the dependence of the NOx production efficiency (molecules of NOx per unit of energy) on the ambient air pressure (e.g. Goldenbaum et al., 1993) favour NOx production in the lower troposphere (below 5 km).

Therefore, it is reason to believe that the NOx produced by lightning gives a C-shaped profile, even if the most efficient NOx-producing cloud to ground (CG) flashes occur within the lower part of the cloud. Secondly, the vertical mixing brings air from the PBL, which can be enriched in NOx and other ozone precursors from surface sources, into the free troposphere. Due to the non-linear ozone chemistry the strong correlation between strong vertical mixing and lightning can change the chemical impact of the emissions significantly (Flatøy and Hov, 1997).

The data given in Price et al. (1997a) only give the vertically integrated distribution of the sources. In this study a constant altitude emission profile of the NOx (in pptv/time) within each thunderstorm (i.e. each deep convective event from the NASA/GISS GCM data) has been assumed. This is equivalent to assuming that the production rate on a mass per volume basis decreases proportionally with pressure.

As discussed above, the lightning emissions in the CTM take place in connection with deep convective events (8-hour average in the meteorological input data). Thus the convective mixing in the CTM will act to redistribute the NOx from lightning. As the current parameterisation of convective mixing in the CTM does not include downdrafts, one could expect that the amount of NOx emitted into the upper troposphere could be overestimated. However, in the real atmosphere the NOx is emitted directly into the region of strong vertical mixing while in the model the implicit assumption is that it is equally distributed over the whole model layer. This would tend to underestimate the fraction of the NOx reaching the upper troposphere. To what extent these two factors balance each other and how they affect the upper tropospheric NOx budget is difficult to determine with our current CTM.

The vertical redistribution and the extended lifetime of NOx at higher altitudes give a maximum in the contribution of NOx from lightning in the upper troposphere (Figure 2). In July the contribution is strongest in the northern hemisphere over the continents. At mid-latitudes lightning contributes about 100-150 pptv over the continents, and 50-70 ppt over the oceans. The relative contribution of NOx from lightning is largest (80%) in the middle troposphere (6-7 km altitude) in the tropics mainly due to the very low contribution from other sources to NOx in this region.

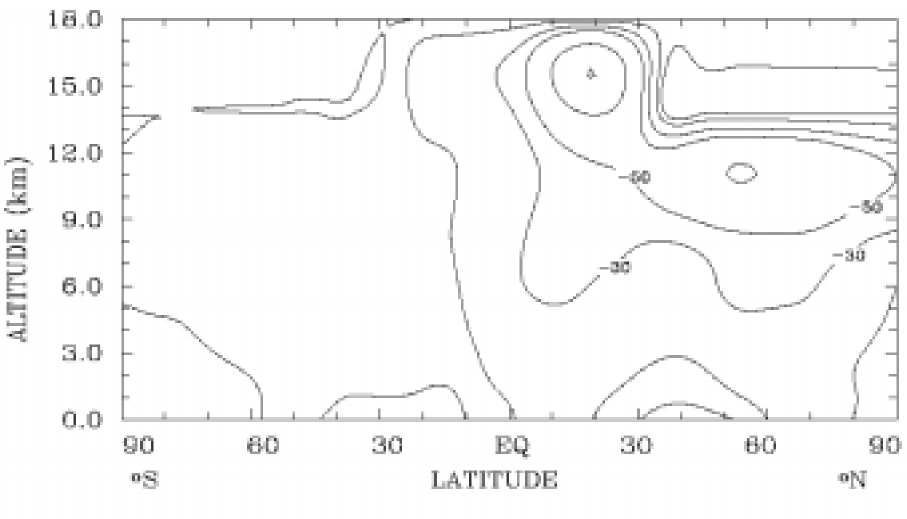

Figure 2. Decrease in zonally averaged NOx concentrations (pptv) in July when lightning sources are omitted. Isolines at 0, -10, -30, -50, -100, and -150 pptv.

4 Results and Discussion

4.1 The reference case

4.1.1 NOx distribution in the upper troposphere

Due to the large latitudinal and longitudinal differences in the source strength of the NOx emissions there is very significant gradients (2-3 orders of magnitude) in the NOx concentrations in the PBL between northern mid-latitude industrialised regions and marine background regions. In the upper troposphere, the chemical lifetime of NOx is longer, and longitudinal mixing is faster. Nevertheless, the CTM calculates significant gradients in the upper tropospheric NOx distribution as well (Figure 3).

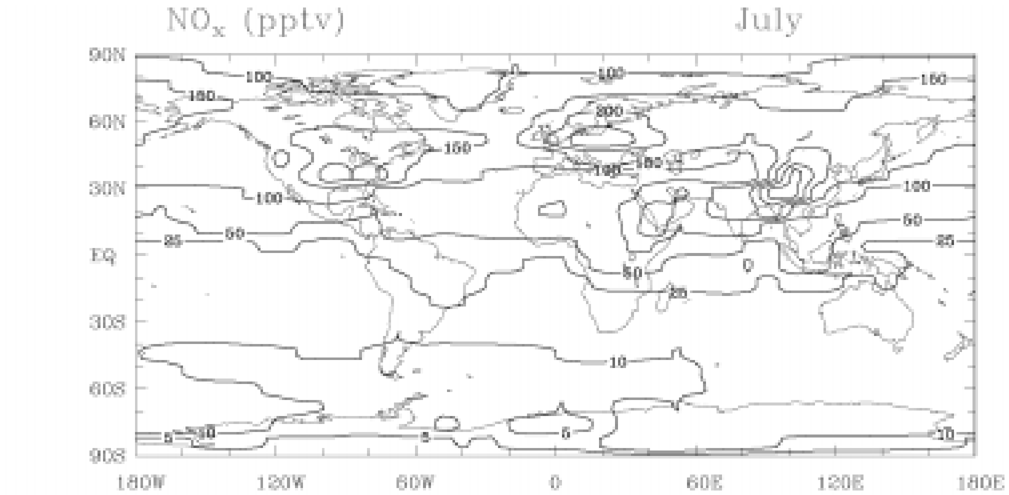

Figure 3. Calculated monthly mean distribution of NOx in case R_0 at approximately 12-km altitude during July. Emissions from aircraft not included. Isolines at 5, 10, 25, 50, 100, 150, 200, 250, and 300 pptv.

At northern mid-latitudes the calculated concentrations of NOx are in good agreement with observations. In the North Atlantic flight corridor (NAFC), NOx measurements taken during POLINAT (Schlager et al., 1997) and the STRATOZ III campaign during June 1984 (Ehhalt and Drummond, 1988) indicated typical NOx daytime levels of 100-300 pptv over the North Atlantic, which is in good agreement with the calculated values. Over the western Pacific (25-42ºN, 10-12 km altitude) Singh et al. (1996a) measured NOx concentrations of 100-150 pptv during September-October 1990.

At lower latitudes measurements during the TRACE A experiment presented by Jacob et al. (1996) indicates NOx-concentrations of the order 50-500 pptv at 9-10 km altitude over Africa, South Atlantic and South America during the biomass burning season in September/October. Using a filtering technique (CO < 80 ppbv) to identify cases not influenced directly by biomass burning TRACE A, Singh et al.(1996b) found NO concentrations of 50-100 pptv during TRACE A Over the tropical Western Pacific (0-20ºN) Singh et al. (1996a) measured 25-50 pptv of NOx in the upper troposphere.

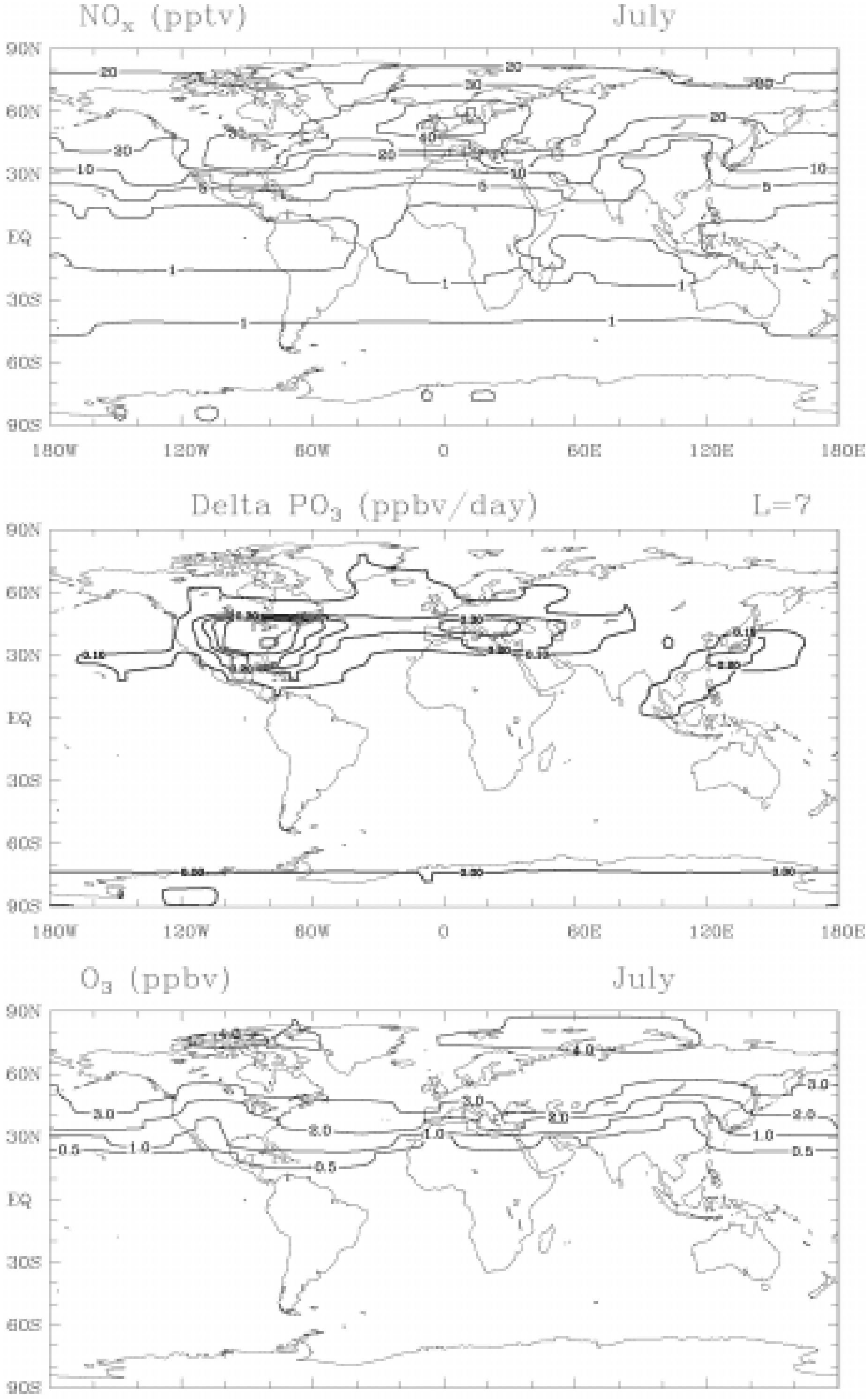

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Figure 4. Calculated increase in NOx (upper panel, . Isolines at 1, 5, 10, 20, 30, 40 and 50 pptv), net ozone production (middle panel, isolines at 0.0, 0.1, 0.2, 0.3, 0.4, 0.4, 0.5 and 0.6 ppbv/day), and ozone concentration (lower panel, ppbv) at 200 hPa in the CTM due to NOx emissions from aircraft in case *R*.

The agreement between calculated and the limited observational data-set of NOx concentrations in the upper troposphere is generally quite good, and better than earlier experiments with this CTM (Jaffe et al., 1997) in which a lightning source of 5 Tg(N)/yr was applied. This could be an indication that 12 Tg(N)/yr of NOx from lightning is more correct. However, there are some indications that some unknown chemistry can recycle HNO_3 to NOx (Chatfield, 1996; Hauglustaine et al., 1996). This is not included in the CTM.

4.1.2 The upper tropospheric NOx budget

The change in the total amount of NOx in any compartment of the free troposphere, over the course of a month, is small compared to the source and sink terms in the NOx budget due to the short chemical lifetime. The contribution to the upper tropospheric NOx budget from the main source processes is given in table 2. The globe has been divided into 9 regions.

The Arctic is everything north of 56°N, the region between 32 and 56°N is divided into 5 regions; Pacific (157.5°E-122.5°W), North America (122.5-62.5°W), North Atlantic (62.5-12.5°W), Europe (12.5°W-47.5°E), and Asia (47.5-157.5°E). Northern hemisphere tropics are the region between the Equator and 32°N.

Table 2: Upper tropospheric NOx budget (390-150 hPa) for July. All values in Gg(N)/month.

Region	Lightning[2]		Convection		Aircraft	
	Gg(N)/month	Norm[3]	Gg(N)/month	Norm.	Gg(N)/month	Norm.
Arctic	37.2	0.33	1.9	0.016	2.20	0.075
Pacific	2.65	0.06	-0.56	-0.004	1.09	0.095
N.America	37.3	1.00	12.2	0.31	8.65	0.86
N. Atlantic	3.08	0.12	-0.17	-0.006	2.19	0.30
Europe	19.8	0.62	33.9	1.00	8.63	1.00
Asia	42.9	0.80	21.3	0.38	9.16	0.64
NH Tropics[1]	243	0.72	52.3	0.15	10.4	0.11

[1]390-70 hPa, due to the higher tropopause height at low latitudes.
[2]12 Tg(N)/yr from lightning.
[3]Normalised flux (per area) for each source. Region with maximum flux is set equal to 1.0.

Due to the different nature of the terms in table 2, the results must be interpreted with care. The 'Convection' term is the net effect of updrafts and subsidence. Therefore, in a situation where the combination of surface sources and convection gives a C-shaped profile of NOx with altitude (i.e. over the continents), the net effect of convection could be close to zero. Also higher emissions from lightning and aircraft tend to decrease the net contribution from convection, as the loss through subsidence increases.

The difference in the contribution from convection between North America and Europe is caused by the location of the more polluted regions. In North America they are located at the east coast (i.e. at the eastern boundary of the region), while in Europe the strongest sources are found to the west. The average concentration of NOx in the PBL is therefore higher over Europe than over North America due to the prevailing westerly winds.

Except for Europe, lightning is the largest source of upper tropospheric NOx, in all regions. When the source terms are normalised with respect to the area of each region, the contribution from aircraft and convection is strongest over the continental mid-latitude regions. An interesting feature is that in July the contribution from lightning at high latitudes is much higher than convection and aircraft, and reach about 50% of the value obtained for the tropical/sub-tropical region

4.1.3 Changes in NOx, ozone and ozone production

Changes in NOx concentrations, net chemical ozone production and ozone concentrations are shown in figure 4. Due to the relatively short lifetime of NOx, the increase in NOx is mainly confined to the flight corridors. The increase in the NOx concentrations are largest over the eastern part of the NAFC, increasing the calculated NOx concentration by about 50 pptv (35%) in July at 200 hPa. Generally, over the north Atlantic between 45and 60 N, the increase is about 35%. The calculated increase in NOx concentrations are higher in the Arctic region than at lower latitudes even if the source of NOx from aircraft per area is stronger in the tropical/sub-tropical region of the northern hemisphere. This is due to less efficient vertical mixing and longer chemical lifetime of NOx at higher latitudes.

The increase in net ozone production at 200 hPa is peaking over the United States, with a maximum extending eastward over the southern parts of the NAFC (figure 4b). The maximum increase in the net production is generally shifted southwards compared to the maximum in the NOx increase due to dependency of the ozone production on solar radiation, non-linear chemical effects, and probably because more intense convective mixing brings up more reactive hydrocarbons fuelling the ozone production. The combination of these two effects leads to almost twice as high concentrations of hydrogen peroxy radicals (HO_2) over south eastern parts of the United States, than over southern Europe when aircraft emissions of NOx are included.

The increase in the ozone mixing ratio is found to be of about 3 ppbv at 200 hPa in July north of about 50N. The northward shift in the ozone increase relative to the increase in the ozone production is mainly caused by the longer residence time of ozone in the upper troposphere at higher latitudes. Due to the higher convective activity at lower latitudes, the enhanced ozone concentrations at these latitudes will tend to be mixed downward and be lost by surface deposition.

4.1.4 Seasonal cycle in the ozone increase

Due to the seasonal cycle in the production and loss rates of ozone, and the seasonal cycle in the transport pattern in the troposphere at mid and high latitudes, there is a strong seasonal cycle in the calculated impact of increased NOx emissions on upper tropospheric ozone. This is illustrated in figure 5, which shows the maximum increase in upper tropospheric ozone (integrated between 390 and 150 hPa) occurs in spring and early summer (April-June). The seasonal maximum occurs earlier at lower latitudes (April at 40° N) and tends to propagate northwards into the summer.

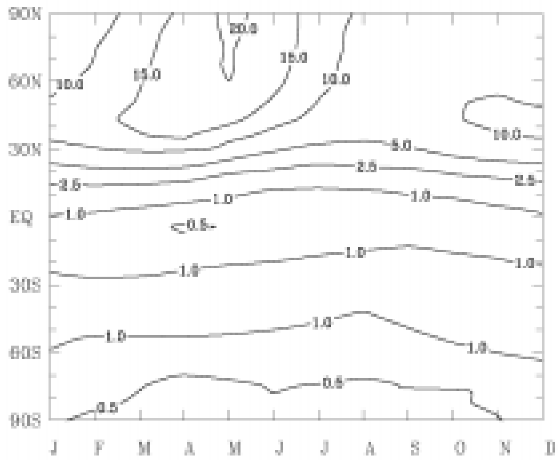


Figure 5. Calculated zonal averaged change in ozone between 390-150 hPa (kg/km^2) due NOx emissions from aircraft for case *R*.

During winter, the increase in ozone is smaller (about a factor of two), and is mainly confined to mid-latitudes, due to the lack of sunlight to drive the ozone production at higher latitudes. The more efficient vertical mixing by convection at low latitudes brings the ozone down towards the surface where it is more readily lost by chemical processes and surface deposition (Jacob et al, 1993). This is causing the minimum in the impact on ozone in the tropics. The increase in ozone becomes slightly higher at mid-latitudes in the southern hemisphere, however, the increase is a factor of 10 stronger at northern mid latitudes, reflecting the large difference in aircraft emissions between the two hemispheres.

4.2 Effects of reduced NOx from lightning and reduced convection

To study the effects of uncertainties in the lightning source and the convective activity we have performed sensitivity studies with reduced lightning emissions (case LL, 5Tg(N)/yr), and reduced convective activity (case LC, convective mass fluxes reduced by 67 %).

As expected reduced emissions of NOx from lightning (case LL) leads to reduced concentrations of NOx, in particular in the upper troposphere (Figure 6). In case *LC* the reduced convection reduce the vertical transport of NOx from the PBL to the upper troposphere. However, the increased residence time of NOx emitted at altitude (mainly lightning, but also from aircraft and by chemical cycling from NOy components) leads to enhanced NOx concentrations. The enhancement of the NOx background is particularly strong in the tropics (about 40% at 12 km altitude), but also at northern mid-latitudes the enhancement reach more than 10%.

The zonally averaged increase in upper tropospheric ozone (390-150 hPa) due to emissions of NOx from aircraft (Figure 7) shows both cases leads to increased effect of NOx from aircraft on ozone compared to case *R*. Reduced emissions from lightning leads to a broader spring/summer maximum at high and mid latitudes (up to 70% higher increase in ozone at 50°N in August/September). At lower latitudes, where the lightning emissions have their maximum, the differences between case *LL* and the reference case is less pronounced. This is due to more intense convective mixing in these regions, which mix down the increased amount of ozone caused by the emissions from aircraft.

The effect of decreasing the convective mixing (case *LC*) is also to increase the enhancement of upper tropospheric ozone due to the emissions of NOx from aircraft at all latitudes and during all seasons. This is due to the prolonged residence time of the ozone produced in the upper troposphere, which more than compensate for the reduced ozone production efficiency caused by the higher NOx background levels.

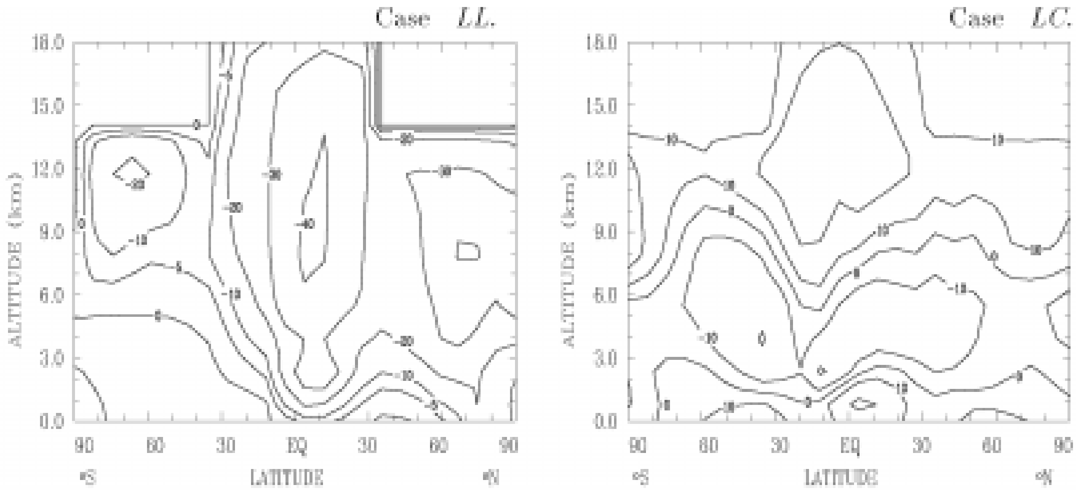


Figure 6. Calculated change (%) in zonal mean NOx mixing ratios in July due to reduced lightning emissions (case R_A versus case LL_A) and reduced convective activity (case R_A versus case LC_A, isolines at -25, -10, 0.0, 10, and 25).

The relative effect is particularly strong in the tropics (150% increase). However, the absolute increase in the upper tropospheric ozone column is only about 30 % of increase in the high latitude region. At northern mid-latitudes the maximum effect occurs during summer, where the increase in ozone is 40-50% higher than in case *R*.

4.2.1 Changes in the upper tropospheric ozone budget

The total integrated amount of ozone in the upper troposphere (390-150 hPa) (Table 3) is strongly affected by the strength of the lightning source and the convection. At northern mid-latitudes we find that the higher lightning source gives 5-7.5% more ozone in the upper troposphere, while decreased convection leads to 15-20% higher ozone amounts compared to case R. The impact of aircraft emissions is fairly similar for all regions at northern mid-latitudes (when normalised to the area of the regions), but varies significantly between the cases (Table 3).

The differences between the three cases are due to a combination of non-linear chemical effects and transport effects in the LC case. As shown in figure 8 all the terms in the ozone budget can change significantly when aircraft emissions are included. In all cases there is an increase in the photochemical ozone production, which is not balanced by an increase in the chemical loss. This leads to an increased vertical gradient between the lower troposphere and the regions directly affected by the aircraft emissions. Thus, the loss due to vertical mixing will increase which is reflected in the convection and vertical advection terms. The approximately equal effect for each region for each case is due to the fast zonal mixing which smooth the differences in ozone production and loss processes between the regions.

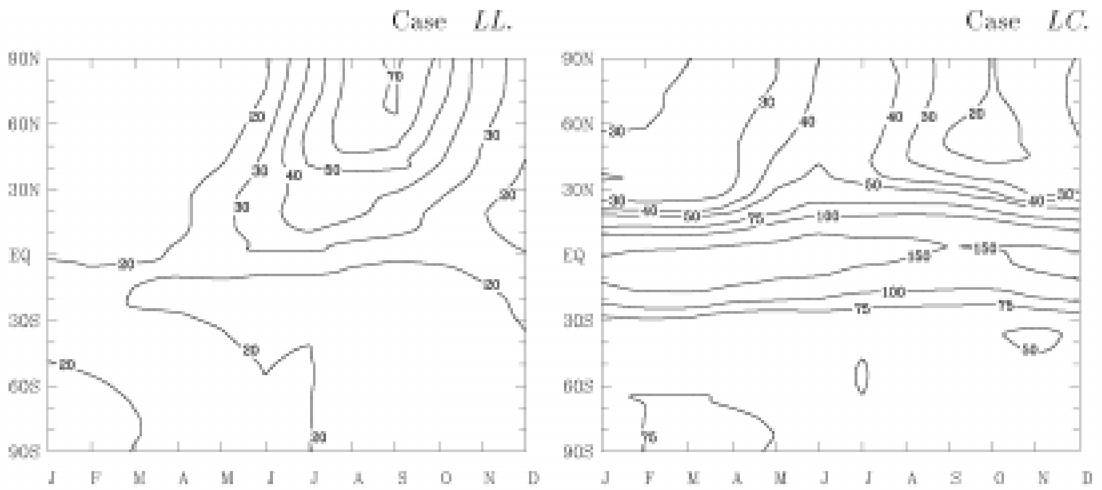


Figure 7. Percentage difference (versus case *R*) in the integrated change in ozone (390-150 hPa) due to emissions from aircraft for cases *LL* (left) and *LC* (right).

A somewhat surprising finding was that the increase in the net chemical ozone production was smallest in case *LC*, in particular over the continental regions of North America and Asia. However, as discussed above, the background NOx levels increase in case *LC* compared to case *R*, which reduce the ozone production efficiency. Compared to case R, the reduced convection also suppresses vertical transport of reactive hydrocarbons from the PBL.

The amount of HO_2 radicals is 30-40% lower at 200 hPa over the continents in case *LC* than in case *R*, making the ozone production less efficient. Compared to case *LL*, the higher emissions from lightning, give higher background NOx levels and therefore the ozone production becomes less efficient. Nevertheless, the increase in ozone concentrations due to emissions of NOx from aircraft, is found to be highest in case *LC*, because the reduced loss of ozone from the upper troposphere by downward mixing more than compensates for the lower production.

Table 3. Integrated upper tropospheric (390-150 hPa) ozone amounts (all numbers in Tg ozone) for July for the three different cases with aircraft emissions. Also shown is the change due to omission of NOx from aircraft in the three cases.

	Case *R*		Case *LL*		Case *LC*	
	Burden	Change	Burden	Change	Burden	Change
Arctic	19.8	-0.62	18.9	-0.83	22.9	-0.91
Pacific	6.67	-0.15	6.16	-0.21	7.67	-0.22
North America	5.31	-0.13	4.96	-0.18	6.23	-0.19
North Atlantic	4.18	-0.12	3.96	-0.16	4.96	-0.17
Europe	5.44	-0.14	5.20	-0.19	6.29	-0.20
Asia	9.02	-0.16	8.59	-0.23	10.3	-0.24
NH tropics[1]	51.8	-0.25	46.5	-0.37	63.7	-0.51

[1]390-70 hPa, due to higher tropopause height in the tropics.

The emission of NOx from aircraft leads to increased ozone production, but also to increased loss from the upper troposphere by transport processes due to steeper vertical gradients in the ozone profile. Also the chemical loss, which is largely proportional to the ozone concentration, is enhanced due to the increase in the ozone concentrations.

4.2.2 Ozone production efficiency

A key question is whether there are pronounced differences between the regions with respect to the ozone production efficiency of aircraft emitted NOx, and the production efficiency depends strongly on the assumptions about lightning and convection activity. This question also relates to how changes in future aircraft emissions, which are likely to cause in increase at lower latitudes, in particular over Asia, will affect the distribution of upper tropospheric ozone.

To address this issue we have calculated a ozone production efficiency (α) given as (for case *R*)

$$\alpha_R = \Delta PO_3(R) / \Delta EM_{NOx}$$

where ΔPO_3 and ΔEM_{NOx} are the increases in chemical ozone production and emissions of NOx from aircraft. Values for α have been calculated for each experiment (α_R , α_{LL} , and α_{LC}) and for each region (Table 4). The α values could be somewhat influenced by transport of aircraft emitted NOx from one region to another, which then will cause ozone production downstream, thereby enhancing the calculated α values in that region. However, the calculated increase in NOx and net ozone production shown in figure 4 indicates that most of the NOx from aircraft is processed in the region where it is emitted.

To study the impact of lightning and convection ozone production enhancements factors
$\beta_{LL} = (\alpha_{LL} - \alpha_R) / \alpha_R$

and

$\beta_{LC} = (\alpha_{LC} - \alpha_R) / \alpha_R$

were also calculated for each region. Higher absolute values of β indicate that the region is specifically sensitive to the assumptions made about the lightning source or the convective activity.

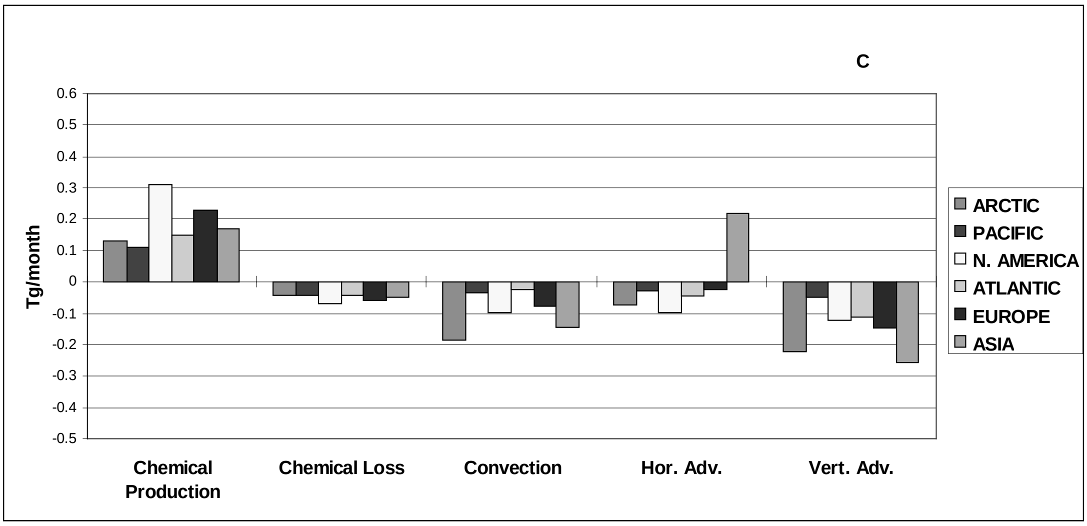
Table 4: Ozone production efficiency, α (kg O_3/kg NOx(N)), and efficiency enhancements, β, in the upper troposphere (390-159 hPa) for the month of July.

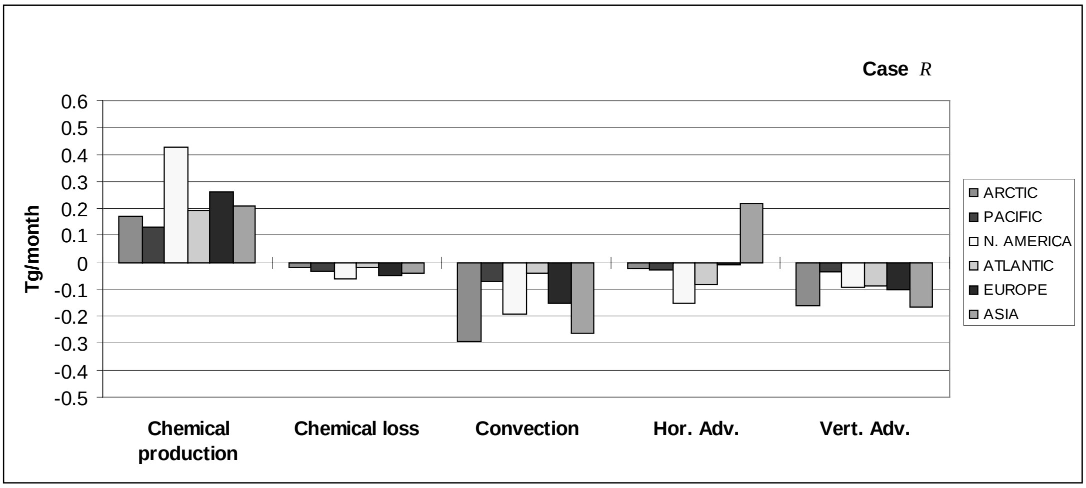
Region	α_R	α_{LL}	α_{LC}	β_{LL}	β_{LC}
Arctic	77.3	144	59.1	0.86	-0.24
Pacific	119	200	101	0.68	-0.15
N. America	49.7	65.4	35.8	0.32	-0.28
N. Atlantic	86.7	128	68.5	0.47	-0.21
Europe	30.1	41.8	26.7	0.39	-0.12
Asia	22.9	34.9	18.6	0.52	-0.19
NH Tropics	86.5	112.5	67.3	0.30	-0.41

As expected the ozone production efficiency is smallest for the continental regions due to the higher background of NOx caused by upward transport of PBL air by deep convection, and higher emissions of NOx from lightning. The highest values for (is found over the Pacific Ocean, where background NOx levels are very low (cf. figure 3). When the lightning source of NOx is reduced to 5 Tg(N)/yr in case LL, the ozone production efficiency increases in all regions as expected.

However, it is somewhat surprising that enhancement of the ozone production efficiency (the β factor) is higher for the marine regions and for the Arctic where the lightning emissions are lower. This can be explained by the coupling of deep convection and lightning emissions in the model, and by the non-linearity of the ozone chemistry. Over the most polluted continental regions (North America and Europe) the elevated NOx levels due to convection means that the relative contribution to the upper tropospheric NOx levels by lightning is smaller. The enhancement of the ozone production efficiency of the NOx emitted from aircraft is therefore smaller in these regions.

The value of β_{LC} is always negative because of the smaller increase in net chemical ozone production in this case, as discussed above. The tropical region appears to be the most sensitive region as convection is more important as a loss of upper tropospheric NOx emitted through lightning in the tropics than in the other regions (cf. figure 6).





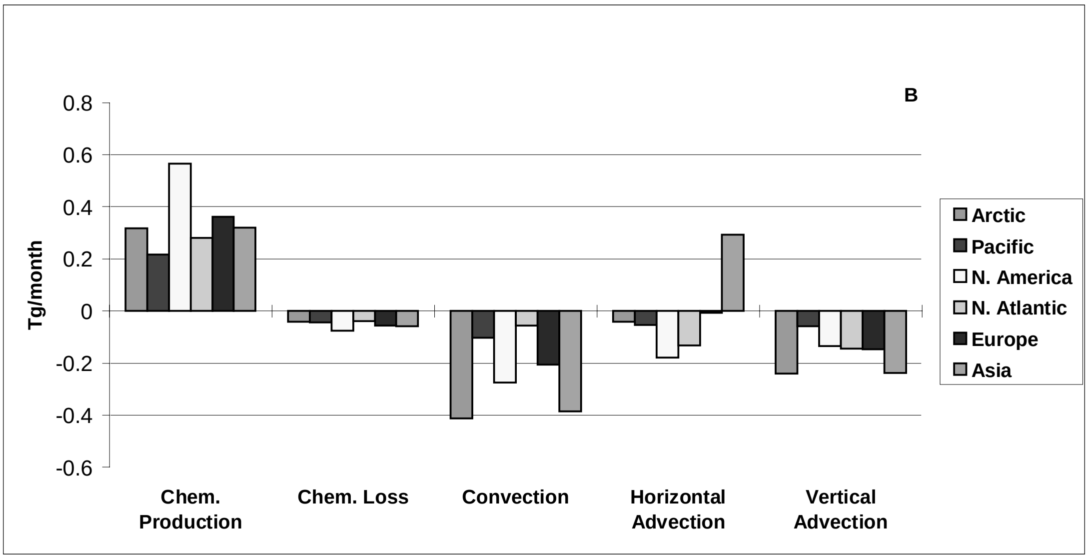


Figure 8. Calculated change in upper tropospheric budget terms (390-150 hPa) for ozone due to inclusion of NOx emissions from aircraft in case R, with lower lightning emissions (LL), and lower convection (LC) cases respectively.

5 Implications for RF Calculations

As discussed above, it was found that the impact on upper tropospheric ozone was very sensitive to the convective activity in the low latitude regions (cf. Figures 5 and 7). In light of the higher climate sensitivity of ozone perturbations at low latitudes this can add up to a significant uncertainty in the global radiative forcing due to aircraft emissions.

A calculation with the University of Reading radiative transfer model adopting a uniform 10 ppbv ozone increase in the troposphere (Berntsen et al., 1997) indicates that the radiative forcing at low latitudes can be as much as a factor of 3 more sensitive to tropospheric ozone changes due to higher surface temperatures, stronger vertical temperature gradients and less clouds at low latitudes.

In addition, estimates of future trends in emissions from aircraft predict a higher growth rate at low latitudes than at mid and high latitudes (Vedontham and Oppenheimer, 1994). It is therefore important to be particularly aware of the tropical regions when assessing the climate impact of ozone increases due emissions from aircraft.

6 Conclusions

A global 3-D CTM has been applied to study the impact of uncertainties in lightning emissions of NOx and deep convective activity on the changes in tropospheric ozone due to NOx emissions from aircraft. A case with reduced emissions of NOx from lightning (5 Tg(N)/yr, case LL) and a case with reduced convective activity (number of events reduced by 67%, case LC) have been compared with the reference case (12 Tg(N)/yr from lightning, case R).

In the reference case we find a zonally homogeneous increase in upper tropospheric ozone concentrations in July of 3-4 ppbv north of 40° Nat 200 hPa altitude. However, the maximum increase is occurring somewhat earlier (April-May), reaching 5-6 ppbv.

In both the perturbation cases (*LL* and *LC*) we find enhanced impact of NOx from aircraft on ozone. The main mechanism in case *LL* is to lower the background levels of NOx in the upper troposphere, thereby increasing the ozone production efficiency per NOx molecule emitted from aircraft. The maximum enhancement (50-70%) in the ozone increase due to NOx from aircraft, compared to the reference case, is found during summer at mid and high latitudes during summer and early fall.

In case *LC* the background concentrations of NOx actually increases because the reduced loss of NOx emitted in the upper tropospheric (mainly from lightning) more than compensate for the reduced transport of NOx from the PBL. The ozone production efficiency of NOx emitted from aircraft is therefore less than in case *R* and *LL*. However, the reduced loss of ozone through transport compensates for this, giving the enhancement of ozone from aircraft in this case.

Uncertainty in the convective activity is found to cause the largest uncertainty in the increase of ozone in the upper troposphere (390-150 hPa), in particular in the tropical region. In the region of maximum ozone perturbations (40°N, spring/summer) both lightning and convection are found to cause about 40% uncertainty in the predicted change of ozone.

7 References

Arnold F., M. Klemm, J. Schneider, V. Bürger, B. Droste-Franke, G. Kirchner, B. Preissler, A. Jung and W. Dann. Trace gas measurements by aircraft based ion molecule mass spectrometry. In. *Pollution from aircraft emissions in the North Atlantic Flight Corridor (POLINAT)*. (edited by Schumann U.) Report EUR 16978 EN, 303 pp. Brussels, Belgium, 1997a.

Berntsen T., and I.S.A. Isaksen. A global 3-D chemical transport model for the troposphere; 1. Model description and CO and ozone results. *J. Geophys. Res.*, 102, 21,239-21,280, 1997.

Berntsen T., I.S.A. Isaksen, J.S. Fuglestvedt, T. Alsvik Larsen, F. Stordal, R.S. Freckleton, and K.P. Shine, Effects of anthropogenic emissions on tropospheric ozone and its radiative forcing, , *J. Geophys. Res.*, 102, 28,101-281, 1997.

Brasseur G.P., J.-F. Müller, and C. Granier. Atmospheric impact of NOx emissions by subsonic aircraft: a three-dimensional model study. *J. Geophys. Res.*, 101, 1423-1428, 1996.

Chatfield, R.B. Anomalous HNO_3/NOx ratio of remote tropospheric air – Conversion of HNO_3 to formic acid and NOx. *Geophys. Res. Lett*, 21, 2705-2708, 1996.

Crutzen, P.J., A discussion of the chemistry of some minor constituents in the stratosphere and troposphere, *Pure Appl. Geophys., 106-108*, 1385-1399, 1973.

Dentener F., and P.J.Crutzen. Reaction of N_2O_5 on tropospheric aerosols: Impact on the global distributions of NOx, O_3 and OH. *J. Geophys. Res.*, 98, 7149-7163, 1993.

Dameris M., Grewe V., Köhler I, Sausen R., C. Bruhl, J.-U. Gross, and B. Steil. Impact of aircraft NOx emissions on tropospheric and stratospheric ozone. Part II: 3-D model results. *Atmosheric Env. In Press*, 1998.

Ehhalt D. H., and J.W. Drummond. NOx sources and the tropospheric distribution of NOx during STRATOZ III. In: *Tropospheric Ozone*, Isaksen, I.S.A. (Ed.). D. Reidel 217-237, 1988.

Flatøy F., and Ø. Hov. NOx from lightning and the calculated chemical composition of the free troposphere. *J. Geophys. Res*., 102, 21,373-21,382, 1997.

Forster, P.M. de F., C.E. Johnson, K.S. Law, J.A. Pyle, and K.P.Shine, Further estimates of radiative forcing due to tropospheric ozone changes, *Geophys. Res. Lett*, 23, 3321-3324, 1996.

Franzbleau E., and C.J. Popp. Nitrogen oxides produced by lightning. *J. Geophys. Res*., 94, 11,089-11,104, 1989.

Fuglestvedt, J.S., I.S.A. Isaksen, and W.-C. Wang, Estimates of indirect global warming potentials for CH_4, CO and NOx, *Clim. Change, 34,* 405-437, 1996.

Goldenbaum G.C., and R.R. Dickerson. Nitric oxide production by lightning discharges. *J. Geophys. Res.,* 98, 18333-18338, 1993.

Hansen, J., G.Russel, D.Rind, P.Stone, A.Lacis, S.Lebedeff, R. Ruedy, and L. Travis, Efficient three dimensional global models for climate studies: Models I and II, *Mon. Weather Rev., 111*, 609-662, 1983.

Hauglustaine D.A., C.Granier, G.P.Brasseur, and G.Megie. The importance of atmospheric chemistry in the calculation of radiative forcing on the climate system. *J. Geophys. Res.,* 99, 1173-1186, 1994.

Hauglustaine D.A., B.A. Ridley, S. Solomon, P.G. Hess and S. Madronich. HNO_3/NOx ratio in the remote troposphere during MLOPEX 2: Evidence for nitric acid reduction on carbonaceous aerosols? *Geophys.Res.Lett.,* 23, 2609-2612, 1996.

Hesstvedt, E., Ø. Hov, and I.S.A. Isaksen, Quasi steady-state approximation in air pollution modeling: Comparison of two numerical schemes for oxidant prediction, *Int. J. Chem. Kinet., X,* 971-994, 1978.

Intergovernmental Panel on Climate Change (IPCC), *Climate Change 1992: The Supplementary Report to The IPCC Scientific Assessment*, edited by J.T. Houghton, B.A. Callander, and S.K. Varney, 200 pp., Cambridge Univ. Press, New York, 1992

Intergovernmental Panel on Climate Change (IPCC), *Climate Change 1995: The Science of Climate Change*, edited by J.T. Houghton et al., 572 pp., Cambridge Univ. Press, New York, 1995.

Isaksen, I.S.A., Ø. Hov, and E. Hesstvedt, Ozone generation over rural areas, *Environ. Sci. Technol., 12,* 1279-1284, 1978.

Jacob D.J. et al. Factors regulating ozone over the United States and its export to the global atmosphere. *J. Geophys. Res*., 98, 14.817-14.826, 1993.

Jacob D.J. et al.. Evaluation and intercomparison of global atmospheric models using ^{222}Rn and other short lived species. *J. Geophys. Res*, 102, 5953-5970. 1997.

Jaffe, D., T.K. Berntsen, and I.S.A. Isaksen, A global three-dimensional chemical transport model for the troposphere, 2, Nitrogen oxides and nonmethane hydrocarbon results, *J. Geophys. Res., 102*, 21,281-21,296, 1997.

Jenkins G. Aviation fuel business, world-wide. Sunningdale Publ. UK, ISBN 1 872546 11 0, 1996.

Kylling A., K.Stamnes and S.-C. Tsay. A reliable and efficient two-stream algorithm for spherical radiative transfer: Documentation of accuracy in realistic layered media. *J. Atmos. Chem*., 21, 115-150, 1995.

Köhler I., R. Sausen and R. Reinberger. Contribution of aircraft emissions to the atmospheric NOx content. *Atmos. Environ*. 31, 1801-1818, 1997.

Lacis A.A., D.J. Wuebbles and J.A. Logan. Radiative forcing of climate by changes in the vertical distribution of ozone. *J. Geophys. Res.*, 95, 9971-9981, 1990.

Lee D.S. et al. Estimations of global NOx emissions and their uncertainties. . *Atmos. Environ*. 31, 1735-1749, 1997.

Lelieveld, J., and R. van Dorland, Ozone chemistry changes in the troposphere and consequent radiative forcing of climate, in *Atmospheric Ozone as a Climate Gas, NATO ASI Ser. I,* vol. 32, edited by W.-C. Wang, and I.S.A. Isaksen, pp. 227-258, Springer-verlag, New York, 1995.

Levine J.S., R.. Rogowski, G.L. Gregory, W.E. Howell, and J. Fishman. Simultaneous measurements of NOx, NO and O_3 production in laboratory discharge: Atmospheric implications. *Geophys. Res. Let.*, 8, 357-360, 1981.

Levy II H., W.J. Moxim and P.S. Kasibhatla. A global three-dimensional time-dependant lightning source of tropospheric NOx. *J. Geophys. Res*., 101, 22911-22922, 1996.

Lin, X., M. Trainer, and S.C. Liu, On the non-linearity of the tropospheric ozone production, *J. Geophys. Res., 93*, 15,879-15,888, 1988.

Müller J.-F. Geographical distribution and seasonal variation of surface emissions and deposition velocities of atmospheric trace gases. *J. Geophys. Res*., 97, 3787-3804, 1992.

Müller J.-F, and G. Brasseur, IMAGES: A three-dimensional chemical transport model of the global troposphere, *J. Geophys. Res., 100*, 16,445-16,490, 1995.

Nüsser H.G. and Scmitt A. Sclussberict zum forschungsvorhaben *Berechung der Emissionsverteilung und deren trends.* BMBF-Förderzeichen: 01 LL 9212/7, 108 pp, 1996.

Prather, M., Numerical advection by conservation of second-order moments, *J. Geophys. Res., 91*, 6671-6681, 1986.

Prather M., M. McElroy, S. Wofsy, G. Russel and D. Rind. Chemistry of the global troposphere: fluorocarbons as tracers of air motion. *J. Geophys. Res*, 92, 6579-6613, 1987.

Price C. and D. Rind. What determines the cloud-to-ground fraction in thunderstorms. *Geophys. Res. Lett*., 20, 463-466, 1993.

Price C., J. Penner and M.Prather. NOx from lightning 1. Global distribution based on lightning physics. *J. Geophys. Res*., 102, 5929-5241, 1997a.

Price C., J. Penner and M.Prather. NOx from lightning 2. Constraints from the global atmospheric circuit. *J. Geophys. Res*., 102, 5943-5251, 1997b.

Roelofs G.J. and Lelieveld J. Distribution and budget of O3 in the troposphere calculated with a chemistry general circulation model. *J. Geophys. Res.*, 100, 20,983-20998, 1995.

Rossow, W.B., and R.A. Schiffer, ISCCP cloud data products, *Bull. Am. Meteorol. Soc., 72*, 2-20, 1991.

Schlager H., P. Schulte, H. Ziereis, P. Konopka, U. Schumann, Ch. Fheigl, R. Marquardt and H. Huntreiser. Aircraft-borne measurements of NOx, O_3 and CO_2. In. *Pollution from aircraft emissions in the North Atlantic Flight Corridor (POLINAT)*. (edited by Schumann U.) Report EUR 16978 EN, 303 pp. Brussels, Belgium, 1997a.

Schumann U. (ed). Pollution from aircraft emissions in the North Atlantic Flight Corridor (POLINAT). Report EUR 16978 EN, 303 pp. Brussels, Belgium, 1997a.

Schumann U. The impact of nitrogen oxide emissions from aircraft upon the atmosphere at flight altitudes - results from the AERONOX project. *Atmos. Environ*. 31, 1723-1734, 1997b.

Singh H.B., M. Kanakidou, P.J. Crutzen and D.J. Jacob. High concentrations and photochemical fate of oxygenated hydrocarbons in the global troposphere. *Nature*, 378, 50-54, 1995.

Singh H.B. et al. Reactive nitrogen and ozone over the western Pacific: Distribution, partitioning, and sources. *J. Geophys. Res.*, 101, 1793-1808, 1996a.

Singh H.B. et al. Impact of biomass burning emissions on the composition of the South Atlantic troposphere: Reactive nitrogen and ozone. *J. Geophys. Res*., 101, 24,203-24,219, 1996b.

Strand A. and Ø.Hov. The impact of man-made and natural NOx emissions on upper tropospheric ozone: A two-dimensional model study. *Atmospheric. Env*. 30, 1291-1303, 1996.

Stevenson D.S., W.J. Collins, C.E. Johnson and R.G. Derwent. The impact of aircraft nitrogen oxide emissions on the tropospheric ozone studied with a 3D Lagrangian model incuding fully diurnal chemistry. *Atmospheric Env*., 31, 1837-1850, 1997.

Vedontham A., and M. Oppenheimer. Aircraft emissions and the global atmosphere; long term scenarios. Environmental Defense Found, New York, 1994.

Wang W.-C. and N.D. Sze. Coupled effects of atmospheric N_2O and O_3 on the Earth's climate. *Nature*, 286, 589-590, 1980.

Wauben W.M.F., P.F.J. van Velthoven, and H.Kelder. A 3D chemistry transport model study of changes in atmospheric ozone due to aircraft NOx emissions. *Atmospheric. Env*., 31, 1819-1836, 1997.

WMO, 1995. Scientific assessment of ozone depletion., Global ozone research and monitoring project. Rep. 37, Geneva, 1995.

Appendix

NOx from lightning

The meteorological input data to the CTM (from the GISS GCM) includes the number of deep convective events every 8'th hour (Ndc), and cloud top height of deep convective clouds (H, in km), averaged over 120 hours. Based on the parameterisation of number of flashes for continental and marine clouds (Fc(H) and Fm(H)) as a function of cloud top height given in Price et al. (1997a), we have calculated the number of flashes for each gridcell in the CTM per 8 hours period. From the emission data from Price et al., zonally averaged emissions for continental (with land-fractions greater than 0.25) and marine regions respectively were calculated (ZPLc).

$$F_c(H) = 3.44 \times 10^{-5} H^{4.92}$$
$$F_m(H) = 6.40 \times 10^{-4} H^{1.73}$$

Similarly, the total number of flashes for continental (FT_c) and marine (FT_m) regions respectively for each latitude band in the CTM was calculated.

$$FT_c = \sum_{m,l} \left[N_{dc} \times F_c(H) \right]$$
$$FT_m = \sum_{m,l} \left[N_{dc} \times F_m(H) \right]$$

Where the sum is over all 8 hour periods during a month, and over continental and marine longitudes respectively. The source strength of NOx from lightning during each 8-hour period in each grid column in the CTM was then calculated by

$$\mathrm{PL}_{\mathrm{NOx}}(\theta, \varphi, t) = N_{dc}(\theta, \varphi, t) \times F_c(H) \times ZPL_c(\varphi, t) / FT_c(\varphi)$$

for continental grid-cells and by

$$\mathrm{PL}_{\mathrm{NOx}}(\theta, \varphi, t) = N_{dc}(\theta, \varphi, t) \times F_m(H) \times ZPL_m(\varphi, t) / FT_m(\varphi)$$

for marine grid-cells.

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