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NO_x Emissions from Aircraft: Its Impact on the Global Distribution of CH_4 and O_3 and on Radiative Forcing

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ABSTRACT

Model studies of the impact of aircraft emissions on atmospheric ozone, and on the methane lifetime have been performed, using a global 3-D CTM (The Oslo CTM1). The calculated changes in the global distribution of ozone and methane have been used to calculate Radiative Forcing (RF) of the current and future fleets (2015 and 2050) of subsonic aircraft. The calculations show that ozone perturbation from aircraft emission occur predominantly in the Northern Hemisphere at mid and high latitudes. Maximum increase in ozone is found in the upper troposphere in the 10 to 12 km height region. The annual average future increases in the region of maximum increase, is estimated to be approximately 12 ppb in 2015. Scenarios for NO_x emissions in 2050 are less certain, however; calculations give a near linear increase in global ozone perturbation for increasing NO_x in the future over the range of NO_x emissions assumed to be realistic until 2050. Calculations with NO_x emissions well beyond this range (50 % higher than the high NO_x emission) showed non-linear ozone response with less ozone increase per NO_x increase than in the cases with lower NO_x emission from aircraft. There is a reduction in the methane lifetime due to enhanced OH from NO_x emissions. The change in the global methane lifetime is estimated to be -1.3 % in 1992 from aircraft emissions, increasing to -3.9 % in 2050. Estimates of RF from CH_4 and O_3 due to aircraft emissions are of opposite sign and of similar magnitude, which makes it difficult to give reliable estimates of the net impact. The regional patterns in RF for the two compounds are however highly different. It is therefore likely that the radiative impact from NO_x emissions could be larger than what can be obtained from global average RFs.

(Key words: Aircraft emissions, Radiative forcing, NO_x, CH_4, O_3)

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1. INTRODUCTION

Aircraft flying at cruising levels at approximately 8 to 13 kms height in the atmosphere affects the atmospheric chemical composition in a height region where the induced changes in the composition could lead to significant changes in climate. This has been demonstrated in two recent international assessments (Brasseur et al. 1998; IPCC 1999). Several greenhouse components that are contained in the aircraft emissions have been identified as potential contributors to upper tropospheric (UT) and lower stratospheric (LS) chemical perturbations, and thereby contribute to changes in radiative forcing (RF). Components, which are expected to be perturbed, on a regional to global scale by aircraft emissions are: CO_2, CH_4, contrails, ozone, and sulphate and soot particles (direct effects). The perturbations of CH_4 and ozone are caused by secondary chemical effects, as a result of NO_x emissions.

In addition changes in ozone levels in the upper troposphere and lower stratosphere could affect the UV radiation penetrating to the lower troposphere through changes in the ozone column densities. Studies of the UV-B impact, performed as part of the assessments (Brasseur et al. 1998; IPCC 1999) indicate that environmental implications of the UV-B changes are less significant than from the changes in RF.

There are two main reasons why aircraft emissions are important. Firstly, air traffic and the emission of pollutants from air traffic have increased rapidly over the last two to three decades, and the prognosis for future emissions indicate continued rapid increases over several decades to come. What is particularly significant is that the increase is expected to be much larger than the general emission of pollutants. This means that we should expect the impact of aircraft emissions on the environment to become more important in the future than it is today. An obvious consequence of this is that good prognoses of future aircraft emissions are essential for performing reliable future estimates of aircraft emissions.

Secondly, emissions occur in height regions (UT and LS) where the impact on ozone and climate could be large. The UT and LS are height regions where lifetimes of gases, like ozone and NO_x compounds, are significantly longer than in the lower and middle troposphere. Emissions of NO_x at these heights will therefore affect the ozone chemistry more efficiently than emissions at surface levels, where gases are broken down more rapidly. In UT meteorological conditions for contrail formation are often favourable for contrail formation. The impact due to perturbations of climate compounds is more efficient at these heights where the temperatures are much lower than in the lower troposphere. The impact due to cloud perturbations is totally different from what is found at lower levels. The results of enhanced cirrus formation and formation of contrails will be an increase in RF. Up to now limited studies of aircraft emissions on cirrus clouds have been performed although significant impacts are possible.

The calculations give contributions of similar magnitude from several climate compounds, i.e., from CO_2, CH_4, contrails, ozone, and significantly lower contributions from sulphate and soot particles (direct effects). This was demonstrated in the recent IPCC (1999) assessment of aircraft impact on the atmosphere (IPCC 1999). However, it was shown that there are uncertainties of such a magnitude that they clearly limit our ability to accurately quantify the contribution from the different compounds. This in turn reduces our possibility to come up with adequate suggestions on how to reduce future aircraft impact.

A large number of model studies of the impact of NO_x emissions from subsonic aircraft have been performed over the last 20 years. During the last few years these studies have been based on 3-D CTMs. Recent assessments of the atmospheric effects of aircraft emissions were completed by NASA (Friedl 1997) and the European Community (Brasseur et al. 1998). For these reports, global studies of the ozone perturbation, due to the present-day aircraft fleet, were performed by several CTM and one GCM models. The model studies used the NASA database in Friedl (1997) and the DLR-2 (Schmitt and Brunner 1997) database in Brasseur et al. (1998). Although there are clear differences in the calculated perturbations caused by aircraft emissions, all model calculations show significant increases in NO_x concentration in the upper troposphere (up to 50% above those calculated without aircraft) in the latitude band where traffic is most frequent (30-60°N). The corresponding increases in ozone concentration in the upper troposphere are up to 15% above those calculated without aircraft. Comparisons revealed that there are significant differences in the calculated ozone perturbations among the models, both in magnitude and in seasonal variation.

In this paper we will focus on a recent study performed as part of the modelling contribution to the IPCC assessment of aircraft impact (IPCC 1999). In the IPCC (1999) assessment study a thorough evaluation of current and future aircraft emissions was performed, with several modelling groups participating in making the predictions and model comparisons of future aircraft impact on the atmosphere.

2 EMISSION SCENARIOS

The estimates focus on three time slices: The current atmosphere (selected as 1992), and two future atmospheric situations: 2015 and 2050. Estimates for the 1992 emissions have been given, as well as projections for 2015 (IPCC 1999). For 2050 estimates have been made based on extrapolations of the 2015 emissions. In addition options exists for high or low growth in energy demand, and possibilities for technology improvements. The table below gives the adopted NO_x emissions for current conditions (1992, 2015), a medium and a high case for 2050 (IPCC 1999).

Table 1. Adopted global aircraft emissions of NOx for selected years.

Year	1992	2015	2050 Medium	2050 High
NO_x emissions Tg (N/yr)	0.50	1.27	2.17	3.46

In the model calculations, the aircraft effluents are put into the models as follows. The gridded fuel burn data (kg fuel/day) are first mapped into the model grid. The amount of material emitted into each grid box, is given by the product of the fuel burn, and the emission index. The emitted material is put into the grid box at each time step at the equivalent rate. By this approach, we ignore the effect of plume processing and assume that the emitted material is instantaneously mixed into the grid box. This is probably an overestimate of the NO_x available

for ozone production, since a fraction will be converted to HNO_3 in the plume. For the subsonic model calculations of the perturbations of oxidants, and through them on the impact on the composition, NO_x is the only aircraft emission considered. Since most models do not calculate the hydrological cycle in the troposphere, water vapour perturbations are not calculated. Sulfur, CO and unburned hydrocarbons are also ignored due to small emissions.

The basic scenarios examine some of the important factors affecting the calculated environmental impact of aircraft. However, a number of uncertainties remain in the treatment of chemical and physical processes that may influence the effects from aircraft emissions. A series of special sensitivity calculations were therefore designed to investigate the most important of the recognised uncertainties. The subsonic aircraft sensitivity scenarios, as described later, examine uncertainties in the background atmosphere, the treatment of upper tropospheric and lower stratospheric chemical and dynamical processes, and the different analyses of aircraft emissions.

In a future atmosphere the background emission from other pollutant sources (e.g., NO_x, CO, NMHC, CH_4) are also expected to increase, leading to a change in atmospheric oxidation which could affect the impact of aircraft emissions on the atmospheric composition. For shorter-lived gases, such as carbon monoxide (CO), nitrogen oxides (NO_x), and non methane hydrocarbons (NMHC), standard boundary conditions for the 1992 cases were used. Values for 2015 and 2050 emissions were obtained by interpolating the IPCC IS92a scenario for current and 2100 values. The adopted increases (in %) are shown in Table 2.

Table 2. Increase in emissions of pollutants (in %) from 1992 to 2015 and to 2050.

Year	Sources	2015	2050
CO	Energy	15	66
	Bio mass burning	9	21
NO_x	Energy	45	107
	Bio mass burning	7	22
NMHC	Energy related sources not isoprene	23	66

Methane is a source for tropospheric ozone and affects the oxidation potential through reactions with OH. Changes in tropospheric chemistry due to methane emissions could be significant in the future, and it could also affect the growth of methane itself through non linear impact. The result of this could be chemical induced growth in climate gases as ozone and in methane itself (Isaksen 1987; Wang et al. 1996). Significant uncertainties exist with regard to future methane growth. The IPCC IS92a scenario (IPCC 1992, 1995) projects significant growth in future methane. However more recent projections (WMO 1999) are smaller than those assumed here. Such slow growth rates are in agreement with observations by Dlugokencky et al. (1998), which indicate that methane levels currently are levelling off. Since future methane mixing ratios depend on atmospheric chemistry in general through the impact

on the OH distribution (reaction R6 below) methane concentrations in the atmosphere will depend on the emission of pollutants like NO_x and CO (Karlsdottir and Isaksen 2000). It is therefore rather uncertain how methane mixing ratios will change in the future. If the current slow trend continues during the next century, with little or no increase in the methane concentration, the increase in background ozone will also be substantially less than calculated in these studies.

We have used the following lower boundary mixing ratios (Table 3) for methane:

Table 3. Methane concentrations (in ppb) used as boundary values.

Year	1992	2015	2050
CH4	1714	2052	2793

No difference in geographical patterns for the future emission changes were assumed in these studies; in other words, the same % increase was assumed to take place in each region between 1992 and 2015 and between 2015 and 2050. This is clearly not correct, since there currently are regions where emissions increase much larger than in other regions (e.g., South-East Asia), and regions where the emissions will be strongly controlled (Europe, North America). In order to examine how significant this assumption in the emission pattern is, a special sensitivity study was performed for 2050 by the 3-D CTM (the UIO CTM1). In this study a geographically different emissions increase was assumed based on a regional growth rate taken from IPCC (1995) and given in IPCC (1999). Such regional differential factors are applied only to energy related sources while emission increases from bio mass burning are the same as in the standard case (Table 2).

For the time slices 1992 and 2015, two model runs were made: A basic scenario with no aircraft emissions, and a scenario with aircraft NO_x emissions added (Table 1). For 2050 a model run with a basic scenario and two aircraft runs with the medium and high scenarios were run (Table 1). The different model runs are summarised in Table 4.

Table 4. Base background scenarios and subsonic aircraft NO_x scenarios used in the global model studies. These scenarios are used to study ozone increases, non linearity in ozone productions from aircraft emissions and the impact on methane lifetime and methane concentrations for future aircraft emissions.

1	A	1992 Base (background atmosphere, no aircraft)
2	B	1992 Base + Aircraft (Chapter 9, NASA 1992)
3	C	2015 Base (background atmosphere, no aircraft)
4	D	2015 Base + Aircraft (Chapter 9, NASA 2015)
5	E	2050 Base (background atmosphere, no aircraft)
6	F	2050 Base + Aircraft (IPCC 1999; Chapter 4, medium case)
7	G	2050 Base + Aircraft (IPCC 1999; Chapter 4, high case)

Key factors in connection with aircraft emissions:

1) Most of the aircraft emission of NO_x is estimated to occur at cruising altitudes, with less emissions at lower altitudes, except for enhanced emissions in the planetary boundary layer during takeoff. IPCC (1999) estimates maximum emissions to occur in the 10 to 12 km height range, with some enhancements also in the 9 to 10 km region. IPCC (1999) estimates the average NO_x emission from aircraft in the 10 to 12 km region to be 5 to 8 times larger than the emission in the 2 to 8 km region per km height interval.
2) Most of the aircraft emissions occur in particular regions which are determined by the main flight corridors. Up to the present time, these emissions basically have been over Europe and the US, and over the North Atlantic along the flight corridor for traffic between Europe and the US. Towards 2015 significant increases in traffic between Europe and South East Asia and between the US and South East Asia are expected to occur.
3) Particularly large emission increases are predicted from future air traffic as shown in Table 1. The NO_x emissions are predicted to increase with approximately a factor 2.5 between 1992 and 2015, and possibly as much as a factor 7 between 1992 and 2050.

The significance of these factors for the ozone is explored by the model studies of current and future aircraft emissions.

3. CHEMICAL PROCESS DETERMINING THE OZONE FORMATION

One of the key questions in connection with studies of future ozone changes from NO_x emissions in general is the non linearity in the ozone forming process, with ozone formation becoming less efficient per NO_x molecule emitted at high NO_x levels. Ozone formation occurs via the following sequence of reactions in the troposphere, and in the lowermost part of the stratosphere:

1) Reactions involving odd nitrogen compounds:

$$NO + HO_2 \rightarrow NO_2 + OH \quad R1$$
$$NO_2 + hv \rightarrow NO + O \quad R2$$
$$O + O_2 + M \rightarrow O_3 + M \quad R3$$

2) Reactions involving carbon monoxide:

$$CO + OH \rightarrow CO_2 + H \quad R4$$
$$H + O_2 + M \rightarrow HO_2 + M \quad R5$$

Through the last sequence HO_2 is reformed to react with NO. The main point here is that nitrogen oxides are cycled through reactions R1 - R2, and therefore, this cycle will not limit the ozone forming potential. However, the formation of the other compound involved in the initial step in the ozone formation, HO_2, requires that CO be oxidized. The number of ozone molecules formed is therefore determined by the amount of CO present. HO_2 molecules can in a similar way be formed through the oxidation of CH_4 and other hydrocarbons. Methane oxidation occurs through the reaction with OH:

$$CH4 + OH \rightarrow CH3 + H2O \quad R6$$

Further oxidation leads to the formation of HO_2 and other peroxy radicals that participate

in the ozone formation process. Similar products are formed when higher hydrocarbons are oxidized.

A key factor in limiting the ozone formation in the atmosphere is the efficiency of NO_x removal from the atmosphere. Several reactions participate in the removal in the atmosphere, but in most of the troposphere NO_x is removed by the reaction of NO_2 with OH:

$$NO_2 + OH + M \rightarrow HNO_3 + M \quad R7$$

Although some of the HNO_3 is oxidized to give back NO_x in the upper troposphere, most of HNO_3 is removed without reforming NO_x. This makes reaction R7 an efficient loss reaction for NO_x in the troposphere, with lifetimes of only a few hours to a day in the lower troposphere, and several days in the upper troposphere. The short lifetime of NO_x in the troposphere means that there are large spatial and temporal variations in the NO_x distribution in the troposphere which are essential to take into account when ozone production is estimated.

There are several important loss reactions for ozone in the global troposphere. The following reactions are the most important:

$$O_3 + HO_2 \rightarrow 2OH + O_2 \quad R8$$

$$O_3 + hv \rightarrow O(^1D) + O_2 \quad R9$$

$$O(^1D) + H_2O \rightarrow OH + OH \quad R10$$

Reactions R1 - R10 are also key reactions in determining OH distribution in the troposphere and the lower stratosphere. The key point here is that increases in ozone and nitrogen oxides enhances the OH distribution through reactions R1 and R10, while enhanced carbon monoxide and methane reduces OH through reactions R4 and R6. Furthermore, reactions with OH (R4 and R6) represent the main loss of CO and methane.

Since the emissions of CO and CH_4 from aircraft have little impact on the atmospheric composition compared to the impact from NO_x emitted from aircraft, the result of NO_x emission from aircraft is to enhance NO_x distribution and thereby the ozone distribution through reactions R1 - R3, and to reduce the CO and CH_4 distribution through the interaction with OH through reactions R4 and R6 respectively. NO_x emissions from aircraft will therefore enhance the abundance of one important greenhouse gas (O_3) and reduce the abundance of another (CH_4). A key point in studies of the climate impact of NO_x emission from aircraft is therefore to determine these opposite effects.

The reactions discussed above are main reactions in the ozone forming process, and in determining the distribution of the hydroxyl radicals in the troposphere. However, there are a large number of additional reactions that will modify the distribution. For instance, several other nitrogen oxide compounds are formed (e.g., PAN, N_2O_5, HO_2NO_2) which affect nitrogen oxidation potential, and the formation and destruction of H_2O_2 affect the OH distribution.

4. CTM ESTIMATES OF GLOBAL OZONE PERTURBATION

Calculated zonal average ozone distribution for 1992 by the 3-D Oslo CTM is shown in Fig. 1. The calculated values are average mixing ratios (in ppb) for January. Of importance for the aircraft impact is the ozone distribution in the upper troposphere and lower stratosphere and the processes that affect the distribution in this region. It is seen that transport from the

lower stratosphere during winter conditions affect Northern Hemispheric ozone distribution over large areas. However, there is a significant gradient in ozone mixing ratios with latitude indicating small effect on low latitude ozone due to transport from high latitudes. The model calculates the low upper tropospheric ozone mixing ratios that have been observed. A similar influx of stratospheric ozone to the Southern Hemisphere is calculated to occur during Southern Hemispheric winter.

Calculated zonal average CO distribution for 1992 is shown in Fig. 2. The values are average mixing ratios (in ppb) for January. The main sources of CO are surface emissions, with some in situ production from methane oxidation (Reaction R6). CO has a chemical lifetime in the troposphere of the order of 2 to 3 months. This long chemical lifetime leads to significant CO transport in the troposphere. This is clearly demonstrated in the figure where it is shown that CO is transported over large distances, particularly in the upper troposphere. Due to the efficient transport of CO, its interaction with the OH chemistry (reactions R4 and R5), and the impact of CO on tropospheric chemistry in general, perturbation of OH in any

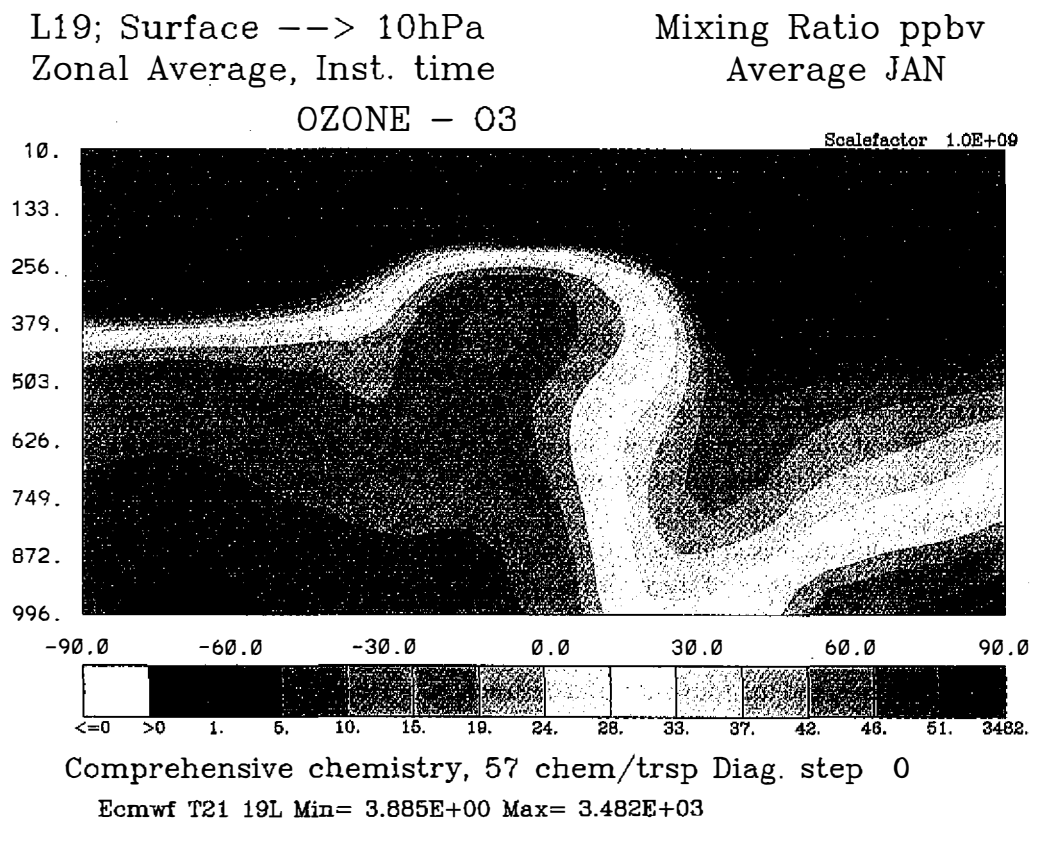

Fig. 1. Zonal and monthly (January) average ozone mixing ratios given in ppb calculated with the Oslo CTM.

limited region of the troposphere will affect on ozone and tropospheric chemistry on a hemispheric scale.

NO_x emissions from aircraft affect tropospheric ozone and the oxidation potential through the sequence of reactions (R1-R3). Calculated ozone perturbations in 2050 from aircraft NO_x emissions (Table 1) using the Oslo CTM are shown in Figs. 3 and 4. Figure 3 presents the zonal average increases of ozone columns in the upper troposphere (between 390 and 150 hPa). The four figures depict ozone increases for different total NO_x emissions and for redistribution of the regional background emissions of pollutants (see figure caption for explanation). Ozone column increases are restricted to middle and high northern latitudes, with little enhancement in the Equator region and in the Southern Hemisphere. The figure shows that the changes in the regional emission pattern for background pollution emission have little effect on the increase in the maximum region as long as the total emission is the same. There are, however, some differences in ozone increases in certain regions where the background levels could be changed significantly in the future (e.g., South-East Asia). One interesting result of the calculations shown in Fig. 3 is that the increase of ozone in the region of max increases

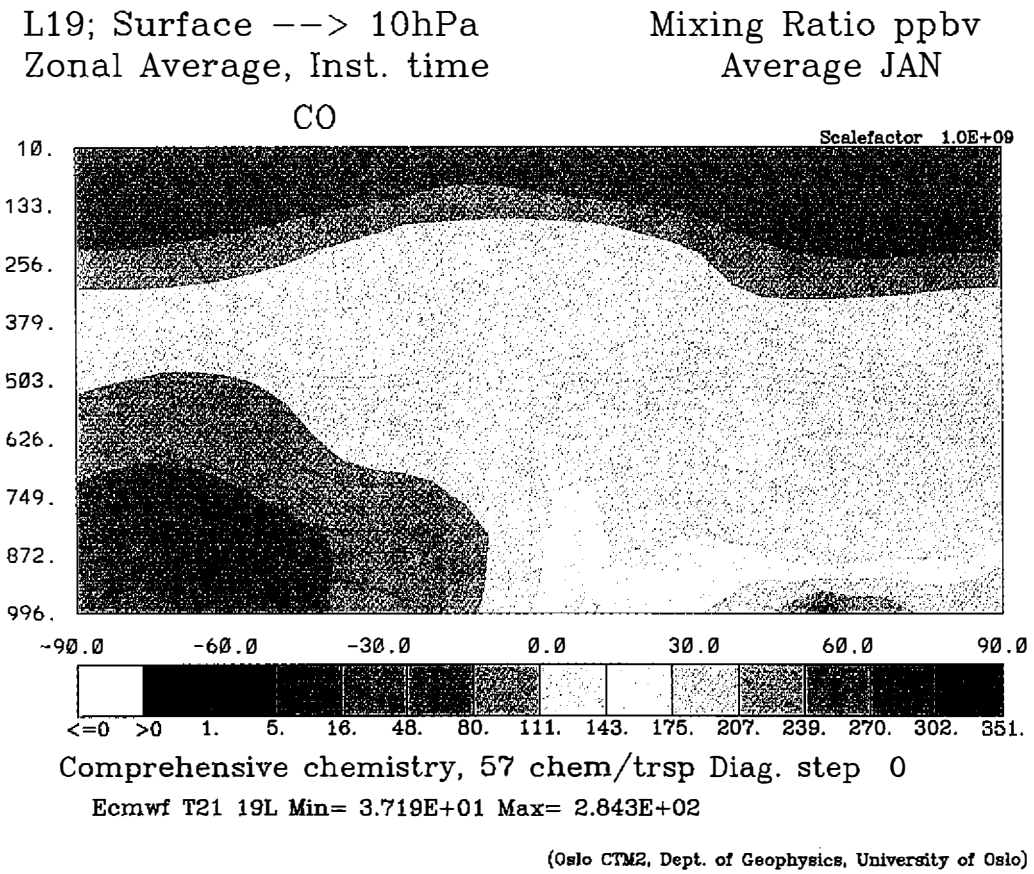

Fig. 2. Zonal and monthly (January) average CO mixing ratios given in ppb calculated with the Oslo CTM.

(high northern latitudes) becomes less pronounced with higher NO_x emissions. This is particularly apparent for the case of extreme high NO_x emission (case G in Table 1 (high) + 50%). The calculated zonal average ozone change, from aircraft emissions (case F in Table 1 (medium)), is depicted in Fig. 4 as monthly averages for January, April, July and October. The ozone increases are restricted to the region of max NO_x emission: upper troposphere/lower stratosphere, mid- to high- northern latitudes. A closer analysis of the model results reveals

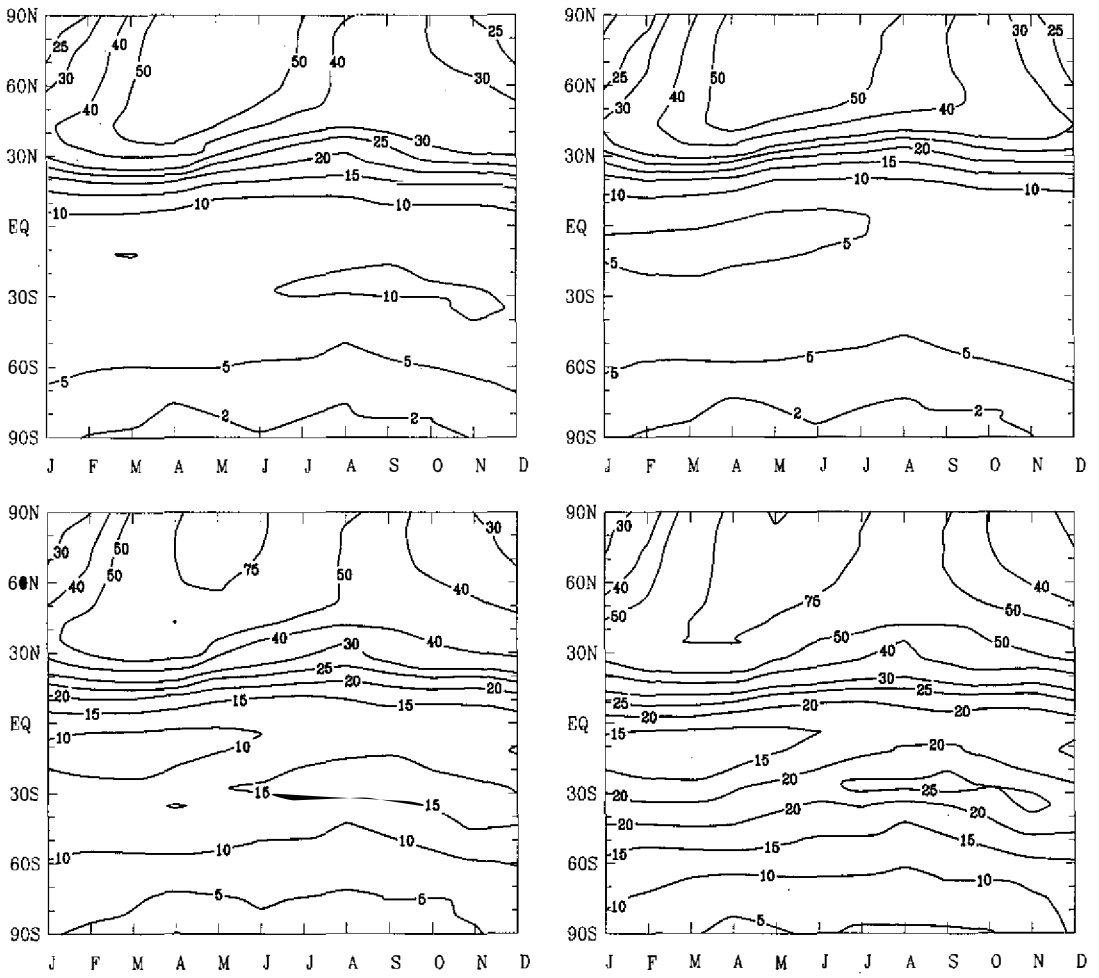

Fig. 3. Calculated zonally averaged change in ozone (kg/km^2) in the upper troposphere (390-150 hPa poleward of 32°, 390-70 hPa at lower latitudes) due to NO_x emissions from aircraft in 2050. Upper left panel case F-E, upper right panel case F-E but with more realistic regional distribution of surface sources, lowr left panel case G-E, and lower right panel case G (further enhanced aircraft emissions by 50%) - case E (for def. of cases, cf. IPCC (1999) page 129.

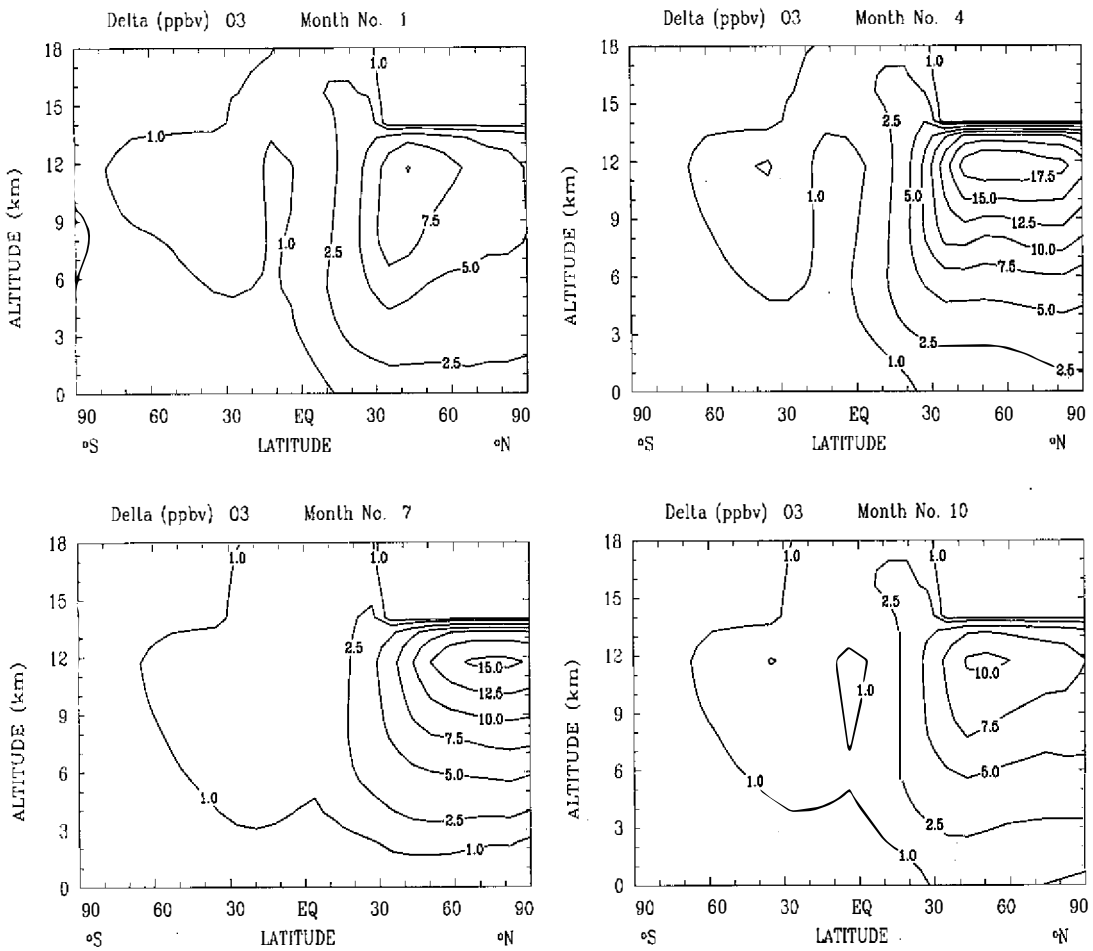

Fig. 4. Calculated zonally averaged change in ozone (ppbv) for January, April, July, and October due to NO_x emissions from aircraft in 2050 for case F-E.

that a significant fraction of the ozone enhancement occurs in the lower stratosphere (25-30%). This is consistent with other model studies which were performed in connection with the IPCC Assessment of aircraft impact on the atmosphere (IPCC 1999). The figure shows that there are large seasonal variations in the ozone perturbations. The largest increases in ozone perturbations are found during spring with a maximum enhancement of approximately 18 ppb. The smallest increases are found during winter with approximately 10 ppb increases in the maximum region. Several factors are believed to contribute to this pattern: Efficiency of ozone production through the reactions R1-R3 (more efficient during the summer months at high latitudes), the background NO_x distribution (more efficient ozone production at low NO_x levels), the efficiency of the transport processes (efficient transport gives less ozone in the region of maximum levels). For instance, a sensitivity study we performed, indicated that half

of the OH impact on the methane lifetime was caused by interaction with the CO chemistry (reaction R4 - R5).

Although the emissions of the precursor NO_x are spatially heterogeneously distributed, the resulting O_3 increases are distributed more uniformly, due to the combined effects of strong longitudinal mixing and the relatively long residence time of ozone in the free troposphere and lower stratosphere. The model shows an efficient transport of excess ozone from the source regions at mid-latitudes to high latitudes, where the residence time of ozone is particularly long due to slower chemistry. This is in agreement with other model studies reported in the 1999 IPCC Assessment (IPCC 1999; Wauben et al. 1997; Stevenson et al. 1997; Berntsen and Isaksen 1999).

A key question when future ozone perturbations from NO_x emissions are studied is the question of non-linearity. Will the ozone production become less efficient per NO_x molecule emitted for high NO_x emissions from aircraft in an environment with higher background levels of NO_x which we will have in the future due to higher surface NO_x emissions? As we have shown in Fig. 3, ozone perturbations are expected to increase significantly in the future with increasing NO_x emissions. A more thorough analysis of the non-linearity in ozone production was done using the results of the Oslo 3-D CTM1 calculations by calculating future (compared to 1992) ozone increases relative to NO_x emissions. The results are given in Table 5.

Table 5. Atmospheric ozone increases from aircraft emission per NO_x molecule emitted, relative to the increase in 1992. The emissions of NO_x are the same as given in Table 1.The values represent increases in global ozone concentrations from the Earth's surface to 16 km obtained with the Oslo 3-D CTM1.

Year	1992	2015	2050	2050 *
Relative ozone increases from NO_x emissions	1.00	0.87	0.81	0.72

* High NO_x emission

The calculations show that there is a gradual reduction in the ozone efficiency for increasing NO_x emissions. There is, however, no dramatic reduction in the efficiency due to the non linearity in the ozone chemistry over the range considered. This was the case both for global ozone and for ozone in the region of maximum increase. However, as we have shown in Fig. 3, if the emissions increase beyond the high emission (case G + 50%), the situation changed significantly. The ozone response to enhanced emission was strongly non-linear. This was particularly apparent in the Northern Hemisphere at mid- and high- latitudes, where most of the emission is assumed to take place. The calculations gave only a 10 to 15 % increase in ozone in the region of maximum increase, for a 50% increase in the NO_x emission. In contrast the ozone increase in the Southern Hemisphere, where the absolute emission increases were much less, was close to the increase in NO_x emission (50 %).

5. INFLUENCE OF NO_x EMISSION ON THE OH DISTRIBUTION AND ON THE CH_4 LIFETIME

The emission of NO_x will lead to higher OH concentrations through reaction R1. This will affect methane lifetime and thereby methane concentrations. In the troposphere, methane is removed mainly by the reaction with the OH radical (Reaction R6). Therefore, a higher OH will lead to more rapid removal of CH_4 from the atmosphere. Table 6 presents the change in chemical lifetime of methane due to aircraft emissions for scenarios 1-6 (Table 4).

Table 6. Changes in methane lifetime due to aircraft emissions based on 3-D CTM calculations. The methane lifetime is defined as the methane amount up to 300 hPa (~10 km) divided by the amount annually destroyed by chemical processes.

Year/Model	1992	2015	2050
Oslo CTM	-1.3	-2.6	-3.9

The perturbation in methane lifetime obtained in this study, is significantly larger than perturbation of methane residence time from aircraft emissions obtained in previous studies (IPCC 1995; Fuglestvedt et al. 1996) using 2-D models. However, calculations made by other CTMs as part of the IPCC Assessment of aircraft impact on the atmosphere, show similar perturbations in the methane lifetime (Isaksen et al. 1999). Methane loss is dominated by OH changes in the tropical and subtropical regions of the lower troposphere. While previous studies showed OH changes that were largely restricted to the upper troposphere, where OH perturbations have little impact on the methane residence time, the current study shows that the perturbations extend well into the lower troposphere at most latitudes in the NH. One explanation for this could be that CO, which accounts for most of the OH loss (Reaction R4), has a sufficiently long lifetime to be transported over large distances. The impact on CO in one region could therefore influence CO and OH in other regions (e.g., low latitude lower troposphere) leading to the estimated impact on methane. The difference in estimated residence time compared with previous 2-D studies could therefore be a result of highly different transport parameterisations in 2-D and 3-D models, with 3-D models having a more efficient transport in the troposphere than the 2-D models.

6. CALCULATIONS OF RADIATIVE FORCING

Both methane and tropospheric ozone are important greenhouse gases. Estimates of their contributions to RF rank them as the 2nd and 3rd most important greenhouse gases affected by man made emissions (after CO_2) since pre industrial time. The most recent IPCC Assessment (IPCC 2001, in preparation) gives RFs since pre-industrial time of 0.48 W/m^2 for CH_4 and 0.35 W/m^2 for tropospheric ozone. Estimates of the stratospheric ozone contribution are negative: 0.15 W/m^2.

We have calculated RF for future changes (2050) in methane and ozone both due to surface emissions of pollutants and due to NO_x emissions from aircraft.

The calculations of RF from surface emissions for CH_4 and O_3 are given in Table 7:

Table 7. Contribution to Radiative Forcing (RF) from future methane, ozone and CO_2 increases.

Time period/Compound	2050-1992	2050-1992*
Methane	0.40	0.13
Ozone	0.31	0.20
Carbon dioxide	1.83	

*Reduced future methane growth as suggested by WMO (1999)

The RF from methane is calculated from the adopted methane increases in 2050 given in Table 3, and from a slower increase given by the WMO Ozone Assessment (WMO 1999). Since ozone is a secondary compound produced by photochemical reactions involving CO, NO_x, CH_4 and NMHC, its future increase will depend on the future increase of all these compounds in the future (Table 2). Although these numbers for RFs are significantly lower than the number given for RF from CO_2, they nevertheless contribute significantly to future RF. Since ozone production is affected by methane, the lower RF for ozone in the second column is a result of lower methane values in this case.

Calculations of RFs for CH_4 and O_3 from aircraft emissions of NO_x are given in Table 8. Ozone gives a positive contribution to radiative forcing, while methane gives a negative contribution to radiative forcing since its lifetime decreases with increasing NO_x emission from aircraft. RF from methane partly cancel out the total RF caused by aircraft NO_x emissions. Also, there are significant uncertainties connected with the calculated changes in methane and ozone concentrations making it difficult to draw firm conclusions concerning the magnitude of the RF from NO_x emissions from aircraft.

The numbers presented in Table 8 are global and yearly average values. The regional patterns for methane and ozone are highly different. Methane is a well-mixed greenhouse gas with perturbations occurring throughout the globe. Contributions to RF come mainly from low and mid latitudes. Ozone, on the other hand, is a short-lived greenhouse gas, with the largest perturbations from aircraft emissions at high northern latitudes, and with small perturbations south of approximately 20 degree N latitudes (Figs. 3 and 4). This results in a contribution to RF almost only from mid and high northern latitudes. At these latitudes the contribution to RF is approximately three times as large as the global average given in Table 8. A similar pattern is seen in the contribution from ozone to RF due to increases in surface emissions. In addition, there are significant seasonal variations in the RF from ozone. We believe therefore the global average numbers given in Table 8 give a too simplified picture of the contribution to radiative impact of aircraft emitted NO_x.

Table 8. Calculated Radiative Forcing (RF) for aircraft emissions given in W/m^2. The numbers are given relative to a model run where there is no aircraft emission. * Gives the result for a model run where different regional growth rates between 1992 and 2050 in background emission are taken into account.

Year/Compound	1992	2015	2050	2050*
Methane	-0.015	-0.032	-0.053	
Ozone	0.020	0.047	0.077	0.068

7. CONCLUSIONS

Aircraft emitted NO_x perturb ozone levels at high northern latitudes with potential for significant increase in the future. The perturbations occur the upper troposphere and lower stratosphere, and are restricted to the latitudes where we have the major flight corridors. A significant part of the ozone perturbation (25-30%) takes place in the lower stratosphere.

Methane is also perturbed by aircraft emissions through the perturbation of the hydroxyl radical (OH). Changes in ozone and CO are important for chemical induced changes over larger regions. Perturbations of these gases in the regions where direct aircraft impact occurs are dissipated to other regions through transport of these gases due to their long lifetimes (months). This affects NO and OH through the reactions R1, R4 and R8.

Although estimates of RF from CH_4 and O_3 due to aircraft emissions are of opposite sign they are of similar magnitude, which makes it difficult to come up with reliable estimates. The regional patterns in RF for the two compounds are highly different. The radiation impact from NO_x emissions could therefore be larger than the what can be obtained from adding the global average RFs.

It is clear that there still are large uncertainties in the estimates of ozone and methane perturbations and in their contributions to radiative forcing. Reduced uncertainties can be obtained through model improvements. An important factor is that the impact of aircraft emissions on the ozone levels takes place in the tropopause region, with a large part of the perturbation occurring in the lower stratosphere. Models used up to now to study the effect of NO_x emissions from aircraft have rather poor representation of the processes occurring in the tropopause region. Furthermore, there is still a significant lack of understanding of ozone loss and production processes in the upper troposphere.

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