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Abstract. A two-dimensional (2-D) model in which dynamics, radiation and chemistry are treated interactively is used to investigate the seasonal changes in tropospheric ozone due to current nitrogen oxide emissions from aircraft and to assess the associated radiative forcing on the climate system. Our results confirm the high efficiency of nitrogen oxide in-situ emissions in producing ozone in comparison to surface emissions. The ozone increase is characterized by a strong seasonal variation; it reaches more than 7 % during summer in the upper troposphere at northen mid-latitudes. On a global average basis, the radiative forcing associated with this ozone increase appears to be small in comparison to that of other greenhouse gases. However, it may play a significant role in the anthropogenic forcing on northern hemisphere climate.

Introduction

Since the late 1970s, several model studies have reported that nitrogen oxide emissions by subsonic aircraft may lead to a significant increase in ozone concentration in the northern hemisphere troposphere [e.g. *Hidalgo and Crutzen*, 1977; *Derwent*, 1982; *Johnston et al.*, 1989; *Beck et al.*, 1992]. Less work has been devoted to the radiative forcing associated with this ozone increase. However, because the O_3 enhancement is most pronounced in the upper troposphere, the radiative forcing can be significant [*Lacis et al.*, 1990]. *Johnson et al.* [1992] have recently estimated the impact on global warming of increases in tropospheric ozone caused by aircraft and surface NO_x emissions using the height-dependent sensitivity of the surface temperature to changes in the ozone profile reported by *Lacis et al.* [1990]. Their results show that the forcing is about thirty times more sensitive to high altitude aircraft emissions of NO_x than to surface emissions. Recently, *Mohnen et al.* [1993] calculated a radiative forcing of 0.04–0.07 W m^{-2} for a prescribed 4–7 % ozone increase between 8 to 12 km and 30°– 50°N.

The purpose of this paper is to investigate the seasonal distribution of the ozone response to current nitrogen oxide emissions by aircraft and to quantify the induced radiative forcing on the climate system. Calculations are performed with a coupled climate-chemistry 2-D model in which *NASA/HSRP* [1993] 1990 aircraft emission estimates have been included.

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The atmospheric model

The model used in this study is a coupled chemical dynamical radiative 2-D model extending from 85°S to 85°N with a latitudinal resolution of 5° and from 0 to 85 km with a vertical resolution of 1 km. The original model has been developed by *Brasseur et al.* [1990] to study the middle atmosphere. This model has been used to investigate the atmospheric effects of stratospheric aircraft [*NASA/HSRP*, 1993; *Tie et al.*, 1994]. Its current version has been extended down to the surface and is described in more detail and validated by *Hauglustaine et al.* [1994a, 1994b]. In this model, the principal surface emission of nitrogen oxides is that from fossil fuel consumption (21 Tg–N/yr). The emissions from soils contribute for 5 Tg–N/yr and biomass burning for 8 Tg–N/yr. The global model surface emission is thus 34 Tg–N/yr, in good agreement with the recent estimate of *Müller* [1992]. The NO_x annual emission from lightning is fixed to an annual mean production of 8 Tg–N and uniformately distributed below 13 km between 60°S and 60°N.

The emission rate of nitrogen oxides by 1990 aircraft fleet is adopted from the *NASA/HSRP* [1993] scenario data sets. Based on this estimate, the global annual consumption of aviation fuel is 1.34×10^{11} kg/yr. The corresponding NO_x total emission for the year 1990 is 1.46 Tg–NO_2/yr (0.44 Tg–N/yr). The NO_x emission distribution peaks in the 30°N–50°N latitude band around 10 km and extends up to 21 km (see *NASA/HSRP* [1993] for more details). In this model, the tropopause height is fixed to an annual mean value ranging from 17 km in equatorial regions to 9 km at high latitudes. About 22 % of the total NO_x emission occur above the model tropospheric domain.

Results

A baseline experiment corresponding to a 1990 background atmosphere in which there are no aircraft emissions is

compared to a simulation in which the aircraft emissions are considered. The calculated NO_y increase is depicted at Figure 1A for January conditions. As obtained by *Hidalgo and Crutzen* [1977] and *Kasibhatla* [1993], the increase shows a dominant poleward direction in the northern hemisphere. A maximum reaching 0.27 ppbv at 11 km is obtained during winter at about 50°N. The relative impact of aircraft emissions in the upper troposphere in the northern hemisphere reaches a maximum of 500 % at 11 km at high latitudes ($\geq$ 70°N). In the upper troposphere (8–12 km) between 30°–60°N our results indicate that aircraft source contributes for about 40–50 % to NO_y. *Hidalgo and Crutzen* [1977] calculated a maximum NO_x increase of 463 % for an NO_x emission of a 2.1 Tg/yr, and *Beck et al.* [1992] obtained a maximum of 100 % in April at 14 km in the latitude band 30°–60°N for a 2.0 Tg/yr emission. *Kasibhatla* [1993] estimates that 30–50 % of the NO_x between 8 and 12 km and between 30°–60°N are from aircraft, and *Ehhalt et al.* [1992] results suggest a contribution up to 40 %. These previous results are consistent with those obtained in this study. Figure 1B illustrates the change in calculated ozone mixing ratio in the troposphere for August conditions when these additional nitrogen oxides emissions are taken into account. As a result of enhanced ozone photochemical production through CH_4 and CO oxidations in the presence of higher NO_x levels, the O_3 mixing ratio increases in the troposphere, specifically in the northern hemisphere where the aircraft effluents are mainly released. It reaches, for exemple, 7 % at 50°N at the altitude of 10 km. The calculated seasonal variation in the maximum ozone increase (at 10 km) is depicted in Figure 1C. The ozone net photochemical production in the troposphere is subject to a strong seasonal variation and peaks during summer when OH, HO_2 and RO_2 radical concentrations and NO_2 photolysis are the largest. As shown in Figure 1C, the calculated ozone increase due to aircraft emissions appears largest in August (7 % at 50°N), but is only of the order of 1–2 % in February. As shown in Figure 1D, the increase in the ozone column abundance exhibits a seasonal variation identical to that shown in Figure 1C, and reaches a maximum of 0.6 % at the end of August. A small ozone decrease associated with emissions occuring in the stratosphere is calculated at high latitudes during spring. The ozone increase calculated with our model is in the range given by previous work and, as far as the seasonal cycle of the perturbation is concerned, is consistent with the results compiled by *Beck et al.* [1992]. The previous model results show maximum increases at around 10 km of 4–12 % in the northern hemisphere depending on the total aircraft NO_x emissions adopted. Note also that, as a consequence of increased O_3 concentrations, the OH abundance increases at 10 km from about 10 % at 30 °N to 40 % at 60 °N.

Table 1 shows the calculated increase in ozone inventory from the baseline experiment for the 1990 NO_x aircraft emissions and for increases in the surface emissions of NO_x and CH_4. In order to illustrate the relation between aircraft emissions and tropospheric ozone production, we adopt the *efficiencies to produce ozone* defined by *WMO* [1991]. This factor provides the ozone increase relative to the emission by aircraft of one mass unit of NO_x, normalized to the impact from methane or surface NO_x. Our model indicates that the NO_x emitted from airplanes in the upper troposphere is about 9 times more efficient in producing ozone than ground based emissions of nitrogen oxides. This feature is a consequence of the higher nitrogen oxide residence time in the upper tro-

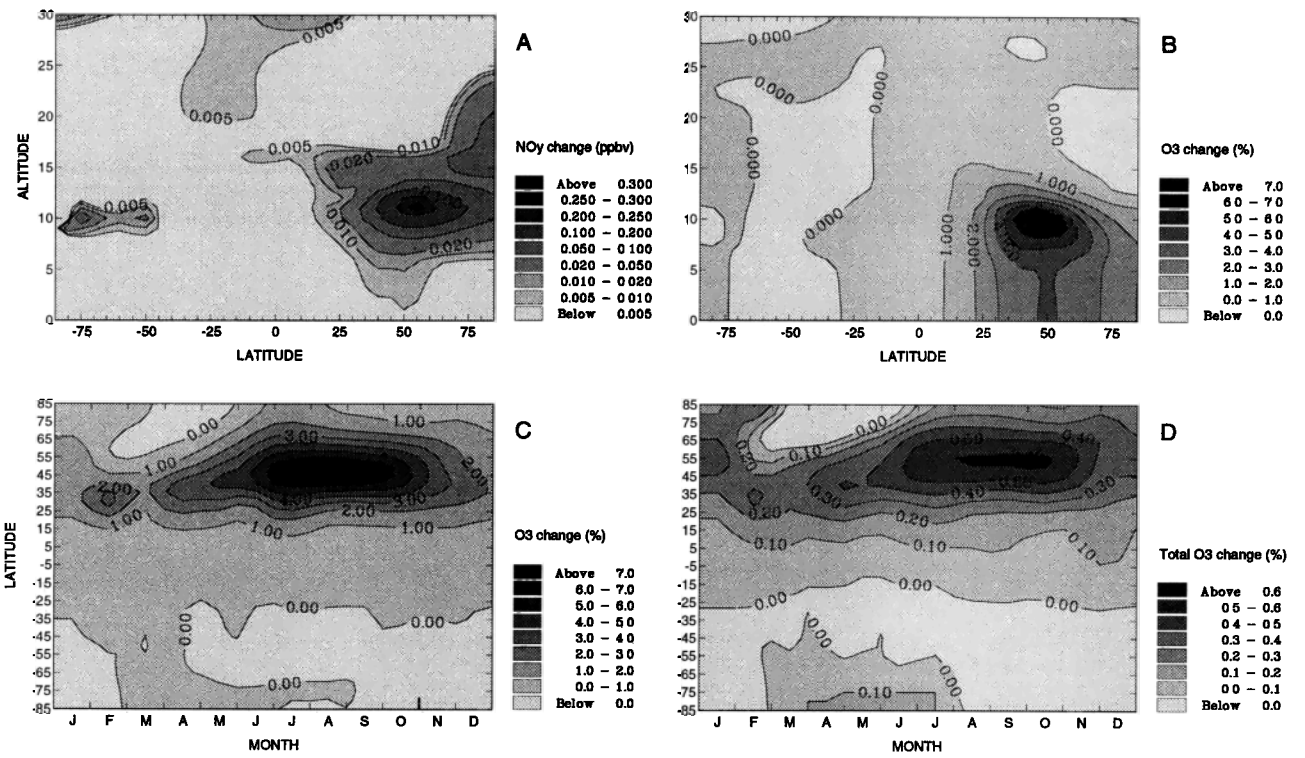

Figure 1. [A] Variation in the NO_y mixing ratio as a function of latitude and altitude for January conditions in ppbv, [B] change in ozone mixing ratio for August conditions in percent, [C] seasonal cycle of O_3 mixing ratio change calculated at 10 km (%), and [D] change in O_3 column abundance as a function of latitude and season.

Table 1. Comparison between changes in emissions ΔE [Tg], calculated increase in tropospheric ozone ΔO_3 [Tg], increase in ozone to changes in emissions ($\Delta O_3/\Delta E$) for methane surface emission doubling, NO_x surface emission doubling and NO_x 1990 aircraft emissions. The *efficiency* is defined as the ozone increase relative to the emission of one mass unit of the constituent and normalized to the impact from methane (column 5) or surface NO_x (column 6) [WMO, 1991].

Case	ΔE	ΔO_3	$\Delta O_3/\Delta E$	Efficiency	
				CH_4	NO_x–surf.
CH_4–surf.	400	37.9	0.09	1	0.16
NO_x–surf.	34	18.8	0.55	6.1	1
NO_x–airc.	0.45	2.33	5.13	57.0	9.3

posphere than in the lower troposphere. As noted by *WMO* [1991], estimates of ozone production resulting from NO_x emissions are highly dependent on the model tropospheric chemical scheme. For exemple, the efficiency of aircraft emissions relative to surface emissions of NO_x reported by *WMO* [1991] was 17 and the study of *Johnson et al.* [1992] provides a value of 22. It should be noted that the formation of oxidants in the troposphere is highly non-linear, implying that the calculated ΔO_3 and $\Delta O_3/\Delta E$ values strongly depend on the assumed increase in NO_x emissions, the background NO_x levels or the lightning emissions [*Beck et al.*, 1992].

We now investigate the radiative forcing on the climate system associated with the calculated increase in tropospheric ozone. The forcing is calculated interactively with the atmospheric composition and is expressed in terms of net radiative budget changes (solar and longwave) at the tropopause. As a consequence of increased longwave trapping by ozone in the lower atmosphere, a positive forcing (downward flux) is calculated. This forcing is 0.015 W m^{-2} on a global and annual mean basis. This value is more than two orders of magnitude lower than the estimated radiative forcing by greenhouse gases since the pre-industrial period [2–2.5 W m^{-2}] [*Hansen and Lacis*, 1990] and even about one order of magnitude lower than the greenhouse forcing over the 1980–1990 period calculated by *Ramaswamy et al.* [1992]. Table 2 compares the globally averaged radiative forcings due to ozone increase (ΔQ) calculated in the case of a doubled surface NO_x emission and 1990 aircraft emissions. Our model suggests that the sensitivity coefficient $\Delta Q/\Delta O_3$ [from the formulation of *Johnson et al.*; 1992] is 1.05 times greater in the case of aircraft emissions than for surface emissions. Since the efficiency to produce ozone is 9.3 times greater in the case of aircraft (Table 1), the sensitivity parameter $\Delta Q/\Delta E$ is 9.8 greater for aircraft than for surface NO_x emissions. These values are lower than that calculated by *Johnson et al.* [1992] with an indirect method based on the *Lacis et al.* [1990] one-dimensional radiative model, giving sensitivity parameters $\Delta Q/\Delta O_3$ and $\Delta Q/\Delta E$ respectively 1.37 and 28.8 times greater for aircraft than for surface emissions. We mainly attibute this discrepancy to the different approches adopted in the two climate forcing calculations. Furthermore, on the basis of the results shown in Figure 1C, the fact that the ozone increase exhibits strong seasonal, latitudinal and vertical variations limits the interpretation that could be made with these globally averaged sensitivity parameters.

Table 2. Comparison between calculated globally averaged net radiative budget change at the tropopause ΔQ [W m^{-2}], change in ΔQ to changes in ozone ($\Delta Q/\Delta O_3$) [W m^{-2}/Tg] and change in ΔQ to changes in emissions ($\Delta Q/\Delta E$) [W m^{-2}/Tg] associated with NO_x surface emission doubling and NO_x 1990 aircraft emissions.

Case	ΔQ	$\Delta Q/\Delta O_3$	$\Delta Q/\Delta E$
NO_x–surf.	0.115	6.12×10^{-3}	0.34×10^{-2}
NO_x–airc.	0.015	6.44×10^{-3}	3.33×10^{-2}

We therefore focus our attention on the seasonal cycle of the ozone forcing associated with aircraft emissions. Figure 2 shows that this radiative forcing exhibits strong seasonal and latitudinal variations. Since the ozone increase reaches a maximum during summer in northern mid-latitudes (Fig. 1C), the radiative perturbation exhibits a maximum during summer in this region of about 0.08 W m^{-2} and decreases to a lower value of about 0.02 W m^{-2} during winter. The enhanced longwave trapping is mainly confined in the northern hemisphere and never exceeds 0.01 W m^{-2} in the southern hemisphere. As a comparison, the 1980–1990 greenhouse forcing associated to all gases and to CFCs only calculated by *Ramaswamy et al.* [1992] are respectively 0.43 and 0.08 W m^{-2} in the 30°N–60°N latitude band. Similarly, with this model, we obtain a 1980–1990 non-CO_2 greenhouse forcing of 0.22 W m^{-2} at 45°N. Furthermore, the model calculated forcings of CH_4, N_2O and CFCs since pre-industrial period are respectively 0.6, 0.3 and 0.15 W m^{-2} in northern mid-latitudes [*Hauglustaine et al.*, 1994a]. The comparison of these results with the maximum forcing illustrated in Figure 2 shows values of the same order of magnitude, stressing the potential importance of aircraft emissions on the local radiative forcing. The summer maximum of 0.08 W m^{-2} for a 7 % ozone increase is in agreement with the forcing indirectly estimated by *Mohnen et al.* [1993]. However, our results stress the strong dependence of the forcing with latitude and season. Note also that, as a consequence of the calculated increase in OH concentration, an indirect radiative forcing is provided by methane concentration changes. Since the major sink of CH_4 is increasing, a negative forcing is predicted. Our results indicate a forcing increasing from -1.4×10^{-3} W m^{-2} at high latitudes to -5.0×10^{-3} W m^{-2} in tropical regions. The globally averaged indirect perturbation, as estimated with this model, is -4.2×10^{-3} W m^{-2}, corresponding to –28 % of the ozone forcing.

Conclusion

The current emission of nitrogen oxides by aircraft may contribute significantly to the perturbation of northern hemisphere tropospheric ozone budget. These model results show that the increase in ozone photochemical production, in presence of higher NO_x levels, leads to enhanced O_3 concentrations, reaching more than 7 % during summer in northern mid-latitude upper troposphere. Our results confirm the high efficiency of nitrogen oxides in-situ emissions in producing ozone in comparison to surface emissions (estimated to be about 9 times more efficient on a global average). The simulated ozone increase is characterized by a strong seasonal variation and ranges from 1–2 % during winter to 7 % during

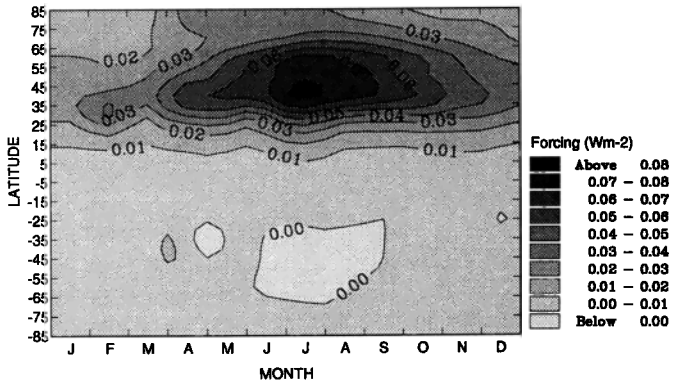

Figure 2. Calculated net radiative forcing at the tropopause [W m^{-2}] due to increased tropospheric ozone mixing ratio represented as a function of latitude and season.

summer at 50 °N in upper troposphere. This result is directly related to the peak in ozone net photochemical production during late spring and summer. A small ozone decrease associated with aircraft emissions occuring in the stratosphere is obtained above the model tropospheric domain. However, this effect is almost offset by the tropospheric ozone increase in the total column change, except at high northern latitudes during spring where a small decrease is predicted.

A particular attention was given to the radiative forcing associated with this increase in tropospheric ozone. The globally averaged positive forcing is only 0.015 W m^{-2}. However, the calculated forcing varies significantly with latitude and season, and reaches 0.08 W m^{-2} in northern mid-latitudes during summer. This maximum value is of the same order of magnitude as the forcing associated with non-CO_2 greenhouse gases during the 1980's and could contribute significantly to the radiative forcing on the northern hemisphere climate. Our results also indicate a globally averaged indirect forcing associated with OH-CH_4 feedback of -4.2×10^{-3} W m^{-2}, corresponding to about –28 % of the tropospheric ozone effect.

It should be noted that several features are ignored in this study. In particular, we investigate the perturbations due to aircraft emissions of nitrogen oxides acting alone. Other aircraft emissions such as those of water vapor, hydrocarbons, sulfur compounds and carbon particles which are neglected in this work could also produce chemical and radiative perturbations. Furthermore, the chemical scheme adopted here does not include NMHC species and the role played by heterogeneous reactions in the troposphere has not been explored. Note also that several studies have stress the limitation of two-dimensional models in assessing both stratospheric and tropospheric chemistry. Despite these uncertainties and limitations, our results suggest that aircraft nitrogen oxide emissions in the troposphere could affect significantly the radiative budget of the surface-troposphere system and need to be further investigated with more detailed chemistry-climate models.

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