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To cite this version:


HAL Id: insu-03860780
https://insu.hal.science/insu-03860780
Submitted on 21 Nov 2022

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Impact of present aircraft emissions of nitrogen oxides on tropospheric ozone and climate forcing

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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 northern 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]. More work has been devoted to the radiative forcing associated with this ozone increase. However, because the O3 enhancement is most pronounced in the upper troposphere, the radiative forcing can be significant [Lacis et al., 1990]. Johnston et al. [1992] have recently estimated the impact on global warming of increases in tropospheric ozone caused by aircraft and surface NOx 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 NOx 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\(^\circ\)–50\(^\circ\)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.

The atmospheric model

The model used in this study is a coupled chemical dynamical radiative 2-D model extending from 85\(^\circ\)S to 85\(^\circ\)N with a latitudinal resolution of 5\(^\circ\) 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 Mailer [1992]. The NOx annual emission from lightning is fixed to an annual mean production of 8 Tg-N and uniformly distributed below 13 km between 60\(^\circ\)S and 60\(^\circ\)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 \times 10^{11} \text{ kg/yr}. The corresponding NOx total emission for the year 1990 is 1.46 Tg–NOx/yr (0.44 Tg–N/yr). The NOx emission distribution peaks in the 30\(^\circ\)N–50\(^\circ\)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 NOx 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 NOx 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 (≥ 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 NOx. Hidalgo and Crutzen [1977] calculated a maximum NOx increase of 463 % for an NOx 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 NOx 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 CH4 and CO oxidations in the presence of higher NOx levels, the O3 mixing ratio increases in the troposphere, specifically in the northern hemisphere where the aircraft effluents are mainly released. It reaches, for example, 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, HO2 and RO2 radical concentrations and NOx 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 occurring 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 NOx emissions adopted. Note also that, as a consequence of increased O3 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 NOx aircraft emissions and for increases in the surface emissions of NOx and CH4. 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 NOx, normalized to the impact from methane or surface NOx. Our model indicates that the NOx 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 tropo-

**Figure 1.** [A] Variation in the NOx 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 O3 mixing ratio change calculated at 10 km (%), and [D] change in O3 column abundance as a function of latitude and season.
The efficiency is defined as the calculated increase in tropospheric ozone \( \Delta O_3 \) [Tg], increase in ozone to changes in emissions \( \Delta \Delta O_3/\Delta E \) for methane surface emissions 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].

<table>
<thead>
<tr>
<th>Case</th>
<th>( \Delta E )</th>
<th>( \Delta O_3 )</th>
<th>( \Delta \Delta O_3/\Delta E )</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH(_4)-surf.</td>
<td>400</td>
<td>37.9</td>
<td>0.09</td>
<td>1</td>
</tr>
<tr>
<td>NO(_2)-surf.</td>
<td>34</td>
<td>18.8</td>
<td>0.55</td>
<td>6.1</td>
</tr>
<tr>
<td>NO(_2)-airc.</td>
<td>0.45</td>
<td>2.33</td>
<td>5.13</td>
<td>57.0</td>
</tr>
<tr>
<td>CH(_4)-airc.</td>
<td>0.45</td>
<td>2.33</td>
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<td>57.0</td>
</tr>
<tr>
<td>NO(_2)-airc.</td>
<td>0.45</td>
<td>2.33</td>
<td>5.13</td>
<td>57.0</td>
</tr>
</tbody>
</table>


Table 1. Comparison between changes in emissions \( \Delta E \) [Tg], calculated increase in tropospheric ozone \( \Delta O_3 \) [Tg], increase in ozone to changes in emissions \( \Delta \Delta O_3/\Delta E \) for methane surface emissions 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].

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\(^\circ\)N–60\(^\circ\)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\(^\circ\)N. Furthermore, the model calculated forcings of CH\(_4\), N\(_2\)O 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 \(-1.4 \times 10^{-3}\) W m\(^{-2}\) at high latitudes to \(-5.0 \times 10^{-3}\) W m\(^{-2}\) in tropical regions. The globally averaged indirect perturbation, as estimated with this model, is \(-4.2 \times 10^{-3}\) 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.

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.

Acknowledgments. Helpful comments by B. Ridley and X. X. Tie are gratefully acknowledged. This work has been supported in part by the CEC. C.G. is supported by the Gas Research Institute. The National Center for Atmospheric Research is sponsored by the National Science Foundation.

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(received 8 September, 1993; revised 24 March, 1994; accepted 12 May, 1994.)