Does the location of aircraft nitrogen oxide emissions affect their climate impact?

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[1] We present results from 112 one-year global chemistry-transport model integrations: a base case, then variants with extra aircraft nitrogen oxide (NOₓ) emissions added to specific regions in the first month (July). The NOₓ stimulates ozone (O₃) production and methane (CH₄) destruction. Responses vary spatially: low background NOₓ regions are most sensitive. Integrated (100-year time horizon) radiative forcings (IRF) are calculated. Net (O₃ + CH₄) IRFs for July aviation NOₓ are generally negative: the global average, weighted by emissions, is −1.9 mWm⁻² yr (TgNOₓ)⁻¹. The positive IRF associated with the short-term O₃ increase (4.1 mWm⁻² yr (TgNOₓ)⁻¹) is overwhelmed by the effects of the long-term CH₄ decrease. Aircraft NOₓ net IRFs are spatially variable, with July values over the remote Pacific approximately balancing the IRF associated with aviation CO₂ emissions (28 mWm⁻² yr (TgNOₓ)⁻¹). The overall climate impact of global aviation is often represented by a simple multiplier for CO₂ emissions. These results show that this is inappropriate.


1. Introduction

[2] The global fleet of aircraft currently account for 2–3% of global anthropogenic CO₂ emissions [Lee et al., 2009], yet these emissions fall outside the remit of the Kyoto Protocol. Between 1970 and 2005 total anthropogenic CO₂ emissions increased by a factor of 2.0; at the same time, aviation CO₂ emissions rose considerably faster (× 2.6) [Lee et al., 2009]. Aircraft emit other climatically important trace species, including oxides of nitrogen (NOₓ = NO + NO₂) and sulphur, water vapour and particulate matter; they also generate contrails [Schumann, 2005] and perturb cirrus clouds [Stordal et al., 2005]. The contribution of aircraft CO₂ towards the radiative forcing (RF) of climate change is relatively well constrained, and is independent of emission location. RFs resulting from the other processes are much less certain, and many of these are dependent on emission location. For example, J. M. Haywood et al. (A case study of the radiative forcing of persistent contrails evolving into contrail-induced cirrus, submitted to Journal of Geophysical Research, 2009), find a large positive localised RF over the North Sea associated with contrail-induced cirrus, however, the spatio-temporal occurrence of such cases, and their global RF, are not well known. Nevertheless, the overall contribution of aircraft to climate change is thought to be larger than that just due to the CO₂ they emit alone [Forster et al., 2006; Lee et al., 2009].

[3] Significant uncertainty surrounds the RF produced by aircraft NOₓ emissions. NOₓ promotes tropospheric ozone (O₃) production, but also stimulates methane (CH₄) destruction by generating hydroxyl radicals (OH) [Wild et al., 2001; Derwent et al., 2001; Köhler et al., 2008]; NOₓ also generates nitrate aerosol and, through changes in oxidants, affects production of other secondary aerosols. These impacts of NOₓ on aerosols, and other interactions between gas-phase and aerosol chemistry are not studied here. Stevenson et al. [2004] (hereafter referred to as ST04) showed that the response of O₃ and CH₄ to aircraft NOₓ is seasonally dependent. Here we extend that analysis to consider how important the emission location is in influencing the impact of aviation NOₓ on O₃ and CH₄.

2. Model and Experiments

[4] We used the STOCHEM global Lagrangian chemistry transport model (CTM), driven by meteorological analyses for 1998 from the Met Office Unified Model. The CTM includes a 70 species tropospheric chemistry scheme and has a horizontal resolution of 5° × 5°, with nine equally spaced vertical levels extending from the surface to ~100 hPa. Full details of the model version employed and the pulse methodology are given elsewhere [Derwent et al., 2008]. Slightly different versions of the same model have been employed in several intercomparisons, and simulated O₃ and OH compare well with other state-of-the-art models and observations [e.g., Stevenson et al., 2006].

[5] A total of 112 integrations were performed; we refer to the first as the base experiment. All integrations ran for one year beginning 1st July, with emissions appropriate for year 2000, including aircraft NOₓ, as used by Dentener et al. [2005]. The 111 variants were all identical to the base except that aircraft NOₓ emissions were increased during the first month of the integration in a particular 10° × 10° region at cruise altitude (~200–300 hPa) by 4 kg (NOₓ) s⁻¹, equivalent to an extra 10 Gg (NOₓ) over the month.

3. Results

[6] We analyse the different responses of CH₄ and O₃ in each experiment, relative to the base (Figure 1). In all cases, a short-lived, regionally distinct, positive O₃ anomaly is produced (Figure 1a), which decays away within a few (<4) months (Figure 1b). The extra NOₓ and O₃ enhance
levels of OH, depleting CH$_4$. A negative CH$_4$ anomaly builds up over the first few months of each experiment, before starting to decay with the 11.5 year CH$_4$ perturbation lifetime (Figure 1c). As CH$_4$ is an O$_3$ precursor, the CH$_4$ depletion is accompanied by a small negative O$_3$ anomaly, which over-rides the effects of the initial positive O$_3$ anomaly within a few months (Figure 1b). Figures 1b and 1c display the perturbations to the O$_3$ and CH$_4$ global burdens for all the experiments, illustrating that aircraft emissions from different locations produce a range of different responses.

[7] The O$_3$ and CH$_4$ anomalies from each experiment have been used to calculate time-integrated radiative forcings (RF). We integrate over a 100-year time horizon, although because the O$_3$ RF changes sign we split it into two components. A time-integrated RF (IRF) is closely related to a Global Warming Potential (GWP), differing only in that it has not been normalized by the IRF from the equivalent mass of CO$_2$ emission [Forster et al., 2007, p. 210]. The initial increase in O$_3$ generates a positive RF with a distinct regional structure, related to both the horizontal distribution of the O$_3$ anomaly (Figure 1a) and the coincident physical properties of the atmosphere and surface (e.g., vertical profiles of temperature and cloud, albedo). Detailed radiative transfer calculations were not performed. Instead, RFs calculated in earlier work (ST04) were normalized to changes in O$_3$ column, for each month and each $5^\circ \times 5^\circ$ model grid-square. These RFs took stratospheric temperature adjustment into account. Changes in O$_3$ column from the new experiments were then converted to grid-square RFs using these fields. Using normalized RFs introduces minor errors associated with slight differences in the vertical profile of the O$_3$ perturbations, but these are insignificant (comparing RFs calculated with the radiative transfer model from ST04 with equivalent RFs calculated from column O$_3$ changes, we find typical differences of less than 5%). For each month, the global RF is calculated from the individual model grid-square RFs. A global, time-integrated RF is then calculated for each experiment by summing all the months with a positive RF. Figure 2a shows the short-term positive O$_3$ IRF (units mW m$^{-2}$ yr) for each of the experiments. Each grid-box in Figure 2
IRFs. To calculate these, we first component (23% of IRF emitted in July, and that different results yr anomaly and the long-term IRF results. The anomaly is globally well- anomaly was 0.23, and we apply this simple scaling here to estimate the long-term O3 IRF components. Figure 2c shows the net IRF arising from both O3 components and CH4 for each experiment.

The short-term O3 IRFs (Figure 2a) are all positive and vary from 1.5 to 11 mW m\(^{-2}\) yr over S. Asia and the S. Pacific, respectively. The CH4 IRFs (Figure 2b) are all negative and vary between —2.8 and —26 mW m\(^{-2}\) yr, with smaller magnitudes over Asia, N. America and Europe, and more strongly negative values over the remote Pacific. The net of the O3 and CH4 IRFs shows a large amount of cancellation, ranging from —22.0 to 0.5 mW m\(^{-2}\) yr, the extremes at locations over the central Pacific and S. America, respectively. It should be noted that these are global average values, and conceal significant regional variation (e.g., the positive O3 IRF will be exerted more locally, see Figure 1a, but the CH4 and long-term O3 IRFs will be exerted more globally), and consequently the climate response may also be quite heterogeneous [cf. Shindell and Faluvegi, 2009].

Spatial variations in the IRF magnitudes correlate well with background NO\(_x\) at the emission location (Figure 3). The largest magnitude O3 and CH4 IRFs tend to be generated from emission sites with low background NO\(_x\), remote from local sources, whilst the lowest magnitude IRFs occur at more polluted locations. Exponential form curves were fit to the data (Figures 3a and 3b); the latitude of emission was also found to be helpful in explaining the O3 IRF results. The following relationships were found:

\[
\text{IRF}_{O3} = 10.7 \exp(-0.0242 \times \text{bNO}_x) - 0.0165 \phi + 2.57; \quad (1)
\]

\[
\text{IRF}_{CH4} = -43.0 \exp(-0.0656 \times \text{bNO}_x) - 3.48; \quad (2)
\]

where background NO\(_x\) mixing ratios (bNO\(_x\)) at the emission sites are in ppt, latitude (\(\phi\)) is in degrees, and the IRFs have units of mW m\(^{-2}\) yr (Tg NO\(_x\))\(^{-1}\). Figure 3c shows the net IRF against background NO\(_x\); this curve is simply the sum:

\[
\text{IRF}_{net} = \text{IRF}_{O3} + 1.23 \times \text{IRF}_{CH4}. \quad (3)
\]

This includes the long-term O3 component (23% of IRF\(_{CH4}\)).

It should be noted that these formulae are appropriate for aircraft NO\(_x\) emitted in July, and that different results would be found for different months, e.g., see ST04. In particular, the latitude dependence probably mainly stems

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**Figure 3.** Scatter plots of IRF components ((a) short-term O3; (b) CH4; and (c) net) against background NO\(_x\) (ppt) at the emission location. Crosses are individual experiments; lines are best-fit curves (see text for details).

represents a global IRF value. The IRFs have been normalized to a pulse of size 1 Tg(NO\(_x\)) to make them directly comparable with earlier work; all IRFs quoted here are normalized to this pulse size.

Figure 2b shows CH4 IRFs. To calculate these, we first extrapolated the CH4 anomaly (Figure 1c) out to 100 years, using our best estimate of the perturbation e-folding time of 11.5 years (see ST04). Because our experiments are only 1 year long, the CH4 anomaly has not had sufficient time to become globally well mixed, consequently it shows some month-to-month variability (Figure 1c), and this causes some uncertainty in the initial value to use for the extrapolation. We use the average of the last eight months of each run to represent the peak CH4 perturbation. We then integrate with respect to time, assume the CH4 anomaly is globally well-mixed, and convert to an IRF using a value of 0.37 mW m\(^{-2}\) ppb\(^{-1}\) (Schimel et al., 1996). There is an additional long-term negative O3 IRF associated with the negative perturbation to the O3 burden (Figure 1b). ST04 found that for pulses of aircraft NO\(_x\) emissions, followed over 5 years, the ratio between the IRF arising from the long-term O3 anomaly and the long-term CH4 anomaly was 0.23, and we apply this simple scaling here to estimate the long-term O3 IRF components. Figure 2c shows the net IRF arising from both O3 components and CH4 for each experiment.

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**Table 1.** Global IRFs (mW m\(^{-2}\) yr (Tg NO\(_x\))\(^{-1}\)) From This Study, Calculated Using Equations (1)–(3), With Comparable Values From Earlier Work

<table>
<thead>
<tr>
<th>Vertical extent and season of pulse, and reference</th>
<th>(\text{IRF}_{O3})/mW m(^{-2}) yr (Tg NO(_x))(^{-1})</th>
<th>(\text{IRF}_{CH4})/mW m(^{-2}) yr (Tg NO(_x))(^{-1})</th>
<th>(\text{IRF}_{net})/mW m(^{-2}) yr (Tg NO(_x))(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cruise altitudes, July, this study</td>
<td>4.1</td>
<td>-4.9</td>
<td>-1.9</td>
</tr>
<tr>
<td>All altitudes, July, (ST04)</td>
<td>5.1</td>
<td>-4.4</td>
<td>-0.26</td>
</tr>
<tr>
<td>All altitudes, Annual [Wild et al., 2001]</td>
<td>7.9</td>
<td>-4.6</td>
<td>+1.8</td>
</tr>
</tbody>
</table>

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from the amount of daylight, which will clearly change with season.

[12] Equations (1)–(3), together with the modelled background NO\textsubscript{x} at 250 hPa and the aircraft NO\textsubscript{x} emission distribution, allow us to construct results equivalent to those reported by ST04 for a July perturbation to aircraft emissions at all locations (Table 1). Ozone and CH\textsubscript{4} IRFs agree to within ~20\% between the two studies, and give an indication of the uncertainty associated with using different versions of the same model. In addition there are slight differences in methodology, such as here we only consider emissions at cruise altitude, whereas ST04 considered all altitudes; the curve-fitting also introduces approximations. Clearly, the net IRF is significantly different, mainly because it is the result of near cancellation of larger terms.

[13] Table 2 presents a breakdown by world region (shown in Figure 1a) of both the emissions and resultant IRFs, based on equations (1)–(3). It is clear that the less polluted regions in general make larger contributions to the IRF components, meaning that emissions in these regions produce more important impacts. In this study, the negative IRFs associated with CH\textsubscript{4} and long-term O\textsubscript{3} reduction almost universally exceed the positive IRFs associated with the short-term O\textsubscript{3} increase. This means the more sensitive regions show more negative IRFs, i.e., the NO\textsubscript{x} emissions in these regions contribute a larger cooling influence. It should be noted that other studies [e.g., Wild et al., 2001] found that the IRF from the short-term O\textsubscript{3} increase dominates the net IRF (Table 1); clearly there is some model-dependence.

### 4. Discussion and Conclusions

[14] Our experiments show that the location of aircraft NO\textsubscript{x} emissions is important, similar to our results for surface NO\textsubscript{x} [Derwent et al., 2008]. A major influence is the background NO\textsubscript{x} at the emission site. More polluted sites are less sensitive to the addition of more NO\textsubscript{x}; the sensitivities of O\textsubscript{3} production and CH\textsubscript{4} destruction also differ. Latitude of emission is also important. We would expect some seasonality in the latitude dependence, based on our earlier results (ST04), but background NO\textsubscript{x} levels appear to be the most important control. Our results reflect the fact that photochemical production of oxidants is non-linearly related to precursor concentrations and UV radiation [Lin et al., 1988; Isaksen et al., 2005]. Grewe and Stenke [2008] simulated the effects of aircraft NO\textsubscript{x} emissions released at 198 hPa in four latitude bands, and found the O\textsubscript{3} RF varied from 18 mW m\textsuperscript{–2} (Tg NO\textsubscript{x})\textsuperscript{–1} in the tropics to 1.5 mW m\textsuperscript{–2} (Tg NO\textsubscript{x})\textsuperscript{–1} at 60–90\°N, similar to our results (Figure 2a).

[15] To put these results in perspective, we can compare them with the associated positive RF (climate warming) from aviation CO\textsubscript{2}. If we take the NO\textsubscript{x} to CO\textsubscript{2} emission ratio from the NASA 1992 inventory [Penner et al., 1999], then the IRF for CO\textsubscript{2} is 28 mW m\textsuperscript{–2} yr for the 1 Tg NO\textsubscript{x} pulse. As CO\textsubscript{2} is long-lived and well-mixed, this value does not vary with emission location or with season, unlike the NO\textsubscript{x} IRF. Locally, the magnitude of the climate cooling from July aircraft NO\textsubscript{x} emissions approaches the IRF from CO\textsubscript{2} (see Figure 2c: ~22 mW m\textsuperscript{–2} yr over the central Pacific). Elsewhere, the positive IRF from CO\textsubscript{2} generally dominates over the negative net IRF from the July NO\textsubscript{x} emissions by a large factor. Whilst we expect some seasonal dependence of our results, we nevertheless expect to find similar results for other months (cf. ST04).

[16] The non-CO\textsubscript{2} climate impacts of global aviation were highlighted by Penner et al. [1999] who introduced the concept of radiative forcing index (RFI), which is the ratio of the total RF at a particular time to the RF derived from CO\textsubscript{2} emissions. Some current policy documents (see Forster et al. [2006] for examples) use an RFI of ~2.5 as a simple multiplicative factor of CO\textsubscript{2} emissions to estimate the climate impact of aviation. Forster et al. [2006] clearly argue that RFI is not an appropriate climate metric for aviation, but that integrated measures of the future effects of an emission, such as IRFs, are much more useful. Net IRFs for aircraft NO\textsubscript{x} calculated here show considerable spatial variation (Table 2). The net IRF over the Atlantic is two-thirds of the value expected based on emissions magnitude alone, whilst the net IRF for the central/southern Pacific is over four times larger than expected. Using a single value to express the climate impact of global aviation NO\textsubscript{x} emissions (or indeed all non-CO\textsubscript{2} effects [e.g., Lee et al., 2009]) therefore conceals considerable spatial variation. The near-cancellation found here between the IRF terms for aviation NO\textsubscript{x} also appears to be model-dependent. Consequently further calculations, across a range of models, of the geographical variations in the non-CO\textsubscript{2} climate impacts of global aviation are urgently required to increase our confidence in these results, and to enable such results to be translated into useful policies.

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### References


### Table 2. Regional Breakdown (%) of Aircraft NO\textsubscript{x} Emissions at Cruise Altitudes and Resulting IRFs

<table>
<thead>
<tr>
<th>Region</th>
<th>NO\textsubscript{x} emission (%)</th>
<th>IRF\textsubscript{O3} (%)</th>
<th>IRF\textsubscript{C16} (%)</th>
<th>IRF\textsubscript{net} (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Trans-Pacific(N)</td>
<td>6.9</td>
<td>10.0</td>
<td>10.0</td>
<td>10.0</td>
</tr>
<tr>
<td>2. N. America</td>
<td>31.0</td>
<td>26.9</td>
<td>25.5</td>
<td>22.6</td>
</tr>
<tr>
<td>3. Trans-Atlantic</td>
<td>9.2</td>
<td>8.8</td>
<td>7.9</td>
<td>6.2</td>
</tr>
<tr>
<td>4. Europe</td>
<td>20.8</td>
<td>14.2</td>
<td>15.4</td>
<td>17.8</td>
</tr>
<tr>
<td>5. Asia</td>
<td>16.2</td>
<td>14.1</td>
<td>13.4</td>
<td>12.1</td>
</tr>
<tr>
<td>6. Trans-Pacific(C/S)</td>
<td>4.1</td>
<td>8.3</td>
<td>11.3</td>
<td>17.6</td>
</tr>
<tr>
<td>7. S. America</td>
<td>3.1</td>
<td>5.1</td>
<td>4.9</td>
<td>4.7</td>
</tr>
<tr>
<td>8. Africa &amp; Mid-East</td>
<td>5.6</td>
<td>6.8</td>
<td>5.6</td>
<td>3.0</td>
</tr>
<tr>
<td>9. Australia</td>
<td>3.1</td>
<td>6.0</td>
<td>6.1</td>
<td>6.1</td>
</tr>
</tbody>
</table>

*Regions (shown in Figure 1a) are arbitrary and have no policy or other significance.*


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