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## 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 ( $\text{NO}_x$ ) emissions added to specific regions in the first month (July). The  $\text{NO}_x$  stimulates ozone ( $\text{O}_3$ ) production and methane ( $\text{CH}_4$ ) destruction. Responses vary spatially: low background  $\text{NO}_x$  regions are most sensitive. Integrated (100-year time horizon) radiative forcings (IRF) are calculated. Net ( $\text{O}_3 + \text{CH}_4$ ) IRFs for July aviation  $\text{NO}_x$  are generally negative: the global average, weighted by emissions, is  $-1.9 \text{ mWm}^{-2} \text{ yr} (\text{TgNO}_2)^{-1}$ . The positive IRF associated with the short-term  $\text{O}_3$  increase ( $4.1 \text{ mWm}^{-2} \text{ yr} (\text{TgNO}_2)^{-1}$ ) is overwhelmed by the effects of the long-term  $\text{CH}_4$  decrease. Aircraft  $\text{NO}_x$  net IRFs are spatially variable, with July values over the remote Pacific approximately balancing the IRF associated with aviation  $\text{CO}_2$  emissions ( $28 \text{ mWm}^{-2} \text{ yr} (\text{TgNO}_2)^{-1}$ ). The overall climate impact of global aviation is often represented by a simple multiplier for  $\text{CO}_2$  emissions. These results show that this is inappropriate. **Citation:** Stevenson, D. S., and R. G. Derwent (2009), Does the location of aircraft nitrogen oxide emissions affect their climate impact?, *Geophys. Res. Lett.*, 36, L17810, doi:10.1029/2009GL039422.

### 1. Introduction

[2] The global fleet of aircraft currently account for 2–3% of global anthropogenic  $\text{CO}_2$  emissions [Lee *et al.*, 2009], yet these emissions fall outside the remit of the Kyoto Protocol. Between 1970 and 2005 total anthropogenic  $\text{CO}_2$  emissions increased by a factor of 2.0; at the same time, aviation  $\text{CO}_2$  emissions rose considerably faster ( $\times 2.6$ ) [Lee *et al.*, 2009]. Aircraft emit other climatically important trace species, including oxides of nitrogen ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) 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  $\text{CO}_2$  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  $\text{CO}_2$  they emit alone [Forster *et al.*, 2006; Lee *et al.*, 2009].

[3] Significant uncertainty surrounds the RF produced by aircraft  $\text{NO}_x$  emissions.  $\text{NO}_x$  promotes tropospheric ozone ( $\text{O}_3$ ) production, but also stimulates methane ( $\text{CH}_4$ ) destruction by generating hydroxyl radicals (OH) [Wild *et al.*, 2001; Derwent *et al.*, 2001; Köhler *et al.*, 2008];  $\text{NO}_x$  also generates nitrate aerosol and, through changes in oxidants, affects production of other secondary aerosols. These impacts of  $\text{NO}_x$  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  $\text{O}_3$  and  $\text{CH}_4$  to aircraft  $\text{NO}_x$  is seasonally dependent. Here we extend that analysis to consider how important the emission location is in influencing the impact of aviation  $\text{NO}_x$  on  $\text{O}_3$  and  $\text{CH}_4$ .

### 2. Model and Experiments

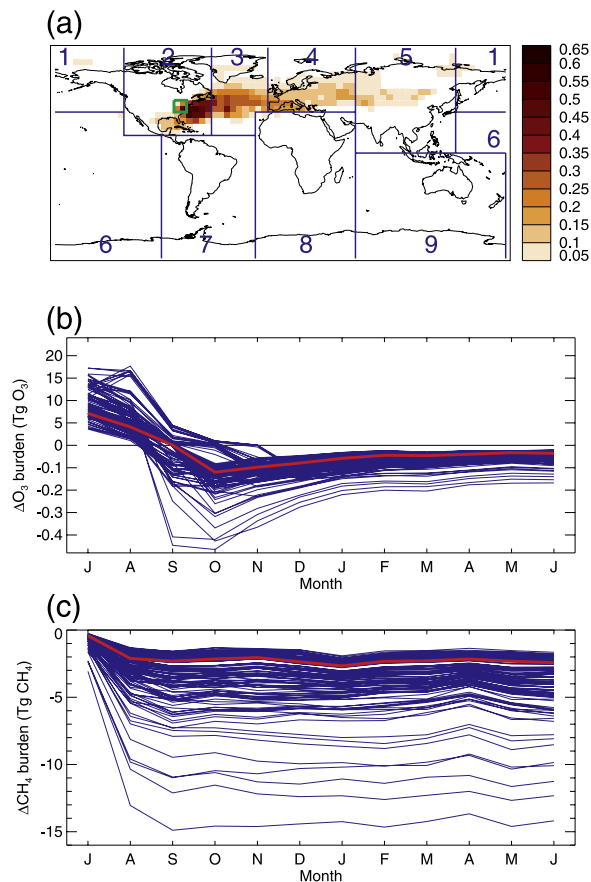
[4] We used the STOCHEM global Lagrangian chemistry transport model (CTM), driven by meteorological analyses for 1998 from the Met Office Unified Model. The CTM includes a 70 species tropospheric chemistry scheme and has a horizontal resolution of  $5^\circ \times 5^\circ$ , with nine equally spaced vertical levels extending from the surface to  $\sim 100$  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  $\text{O}_3$  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  $\text{NO}_x$ , as used by Dentener *et al.* [2005]. The 111 variants were all identical to the base except that aircraft  $\text{NO}_x$  emissions were increased during the first month of the integration in a particular  $10^\circ \times 10^\circ$  region at cruise altitude ( $\sim 200$ – $300$  hPa) by  $4 \text{ kg} (\text{NO}_2) \text{ s}^{-1}$ , equivalent to an extra  $10 \text{ Gg} (\text{NO}_2)$  over the month.

### 3. Results

[6] We analyse the different responses of  $\text{CH}_4$  and  $\text{O}_3$  in each experiment, relative to the base (Figure 1). In all cases, a short-lived, regionally distinct, positive  $\text{O}_3$  anomaly is produced (Figure 1a), which decays away within a few ( $<4$ ) months (Figure 1b). The extra  $\text{NO}_x$  and  $\text{O}_3$  enhance

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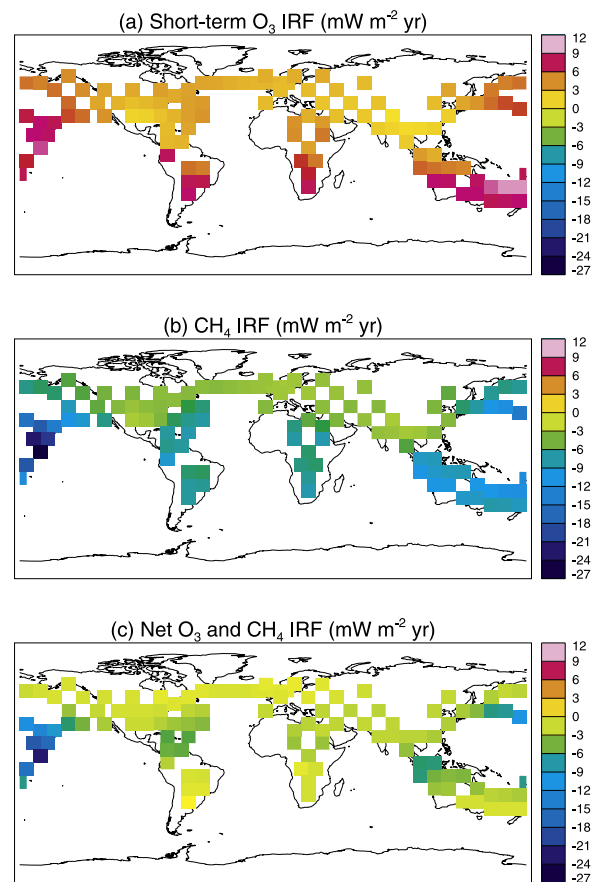


**Figure 1.** (a) The monthly mean change in ozone (ppb) at 250 hPa for July (the month of extra emissions), for an experiment releasing extra aircraft NO<sub>x</sub> over eastern N. America (green box), relative to the base case. The numbered regions are referred to in Table 2. (b) Change in tropospheric ozone burden (Tg) (thick red line) for the 12 months from July. NB the negative scale has been expanded by a factor of 50 for clarity. The thin blue lines show the change for all the other experiments. (c) Same as Figure 1b but for methane. Results in Figure 1a are for the pulse magnitude used in the experiment (10 Gg(NO<sub>2</sub>)), whilst Figures 1b and 1c show results normalised to a 1 Tg(NO<sub>2</sub>) pulse.

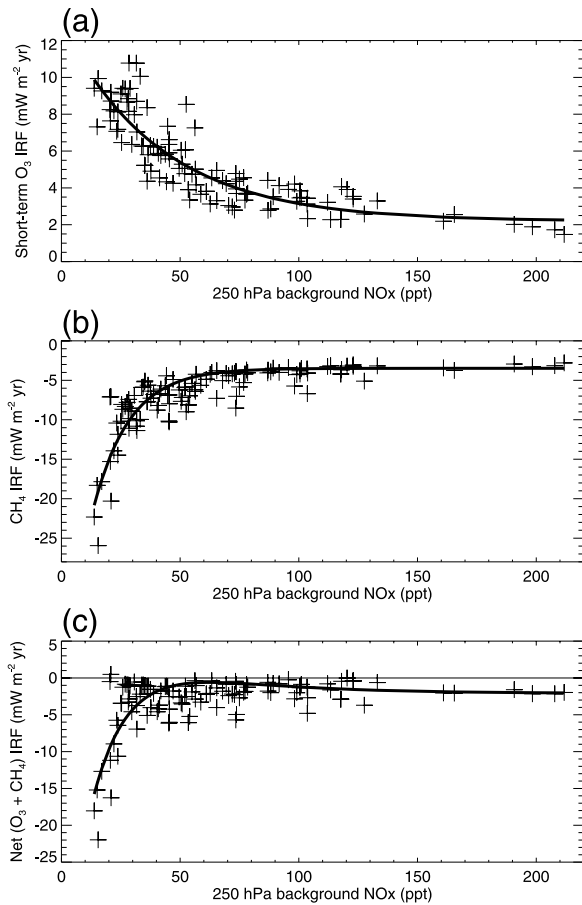
levels of OH, depleting CH<sub>4</sub>. A negative CH<sub>4</sub> anomaly builds up over the first few months of each experiment, before starting to decay with the 11.5 year CH<sub>4</sub> perturbation lifetime (Figure 1c). As CH<sub>4</sub> is an O<sub>3</sub> precursor, the CH<sub>4</sub> depletion is accompanied by a small negative O<sub>3</sub> anomaly, which overrides the effects of the initial positive O<sub>3</sub> anomaly within a few months (Figure 1b). Figures 1b and 1c display the perturbations to the O<sub>3</sub> and CH<sub>4</sub> global burdens for all the experiments, illustrating that aircraft emissions from different locations produce a range of different responses.

[7] The O<sub>3</sub> and CH<sub>4</sub> 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<sub>3</sub> 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<sub>2</sub> emission [Forster *et al.*, 2007, p. 210]. The initial increase in O<sub>3</sub> generates a positive RF with a distinct regional structure, related to both the horizontal distribution of the O<sub>3</sub> 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<sub>3</sub> column, for each month and each 5° × 5° model grid-square. These RFs took stratospheric temperature adjustment into account. Changes in O<sub>3</sub> 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<sub>3</sub> perturbations, but these are insignificant (comparing RFs calculated with the radiative transfer model from ST04 with equivalent RFs calculated from column O<sub>3</sub> 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<sub>3</sub> IRF (units mW m<sup>-2</sup> yr) for each of the experiments. Each grid-box in Figure 2



**Figure 2.** Calculated (stratospheric adjusted) time-integrated radiative forcings (relative to the base) for: (a) the short-term O<sub>3</sub> component; (b) the CH<sub>4</sub> component; and (c) the total net (sum of the short- and long-term O<sub>3</sub> and CH<sub>4</sub> components). Each box shows the global IRF associated with the aircraft emissions from that location. All forcings are normalised to a 1 Tg(NO<sub>2</sub>) emission pulse.



**Figure 3.** Scatter plots of IRF components ((a) short-term O<sub>3</sub>; (b) CH<sub>4</sub>; and (c) net) against background NO<sub>x</sub> (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<sub>2</sub>) to make them directly comparable with earlier work; all IRFs quoted here are normalized to this pulse size.

[8] Figure 2b shows CH<sub>4</sub> IRFs. To calculate these, we first extrapolated the CH<sub>4</sub> 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 CH<sub>4</sub> 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 CH<sub>4</sub> perturbation. We then integrate with respect to time, assume the CH<sub>4</sub> anomaly is globally well-mixed, and convert to an IRF using a value of 0.37 mW m<sup>-2</sup> ppb<sup>-1</sup> [Schimel *et al.*, 1996]. There is an additional long-term

negative O<sub>3</sub> IRF associated with the negative CH<sub>4</sub> anomaly; this is seen in the negative perturbation to the O<sub>3</sub> burden (Figure 1b). ST04 found that for pulses of aircraft NO<sub>x</sub> emissions, followed over 5 years, the ratio between the IRF arising from the long-term O<sub>3</sub> anomaly and the long-term CH<sub>4</sub> anomaly was 0.23, and we apply this simple scaling here to estimate the long-term O<sub>3</sub> IRF components. Figure 2c shows the net IRF arising from both O<sub>3</sub> components and CH<sub>4</sub> for each experiment.

[9] The short-term O<sub>3</sub> IRFs (Figure 2a) are all positive and vary from 1.5 to 11 mW m<sup>-2</sup> yr over S. Asia and the S. Pacific, respectively. The CH<sub>4</sub> IRFs (Figure 2b) are all negative and vary between -2.8 and -26 mW m<sup>-2</sup> yr, with smaller magnitudes over Asia, N. America and Europe, and more strongly negative values over the remote Pacific. The net of the O<sub>3</sub> and CH<sub>4</sub> IRFs shows a large amount of cancellation, ranging from -22.0 to 0.5 mW m<sup>-2</sup> 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 O<sub>3</sub> IRF will be exerted more locally, see Figure 1a, but the CH<sub>4</sub> and long-term O<sub>3</sub> IRFs will be exerted more globally), and consequently the climate response may also be quite heterogeneous [cf. Shindell and Faluvegi, 2009].

[10] Spatial variations in the IRF magnitudes correlate well with background NO<sub>x</sub> at the emission location (Figure 3). The largest magnitude O<sub>3</sub> and CH<sub>4</sub> IRFs tend to be generated from emission sites with low background NO<sub>x</sub>, 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 O<sub>3</sub> IRF results. The following relationships were found:

$$\text{IRF}_{\text{O}_3} = 10.7 \exp(-0.0242 \text{ bNO}_x) - 0.0165\phi + 2.57; \quad (1)$$

$$\text{IRF}_{\text{CH}_4} = -43.0 \exp(-0.0656 \text{ bNO}_x) - 3.48; \quad (2)$$

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

$$\text{IRF}_{\text{net}} = \text{IRF}_{\text{O}_3} + 1.23 \text{ IRF}_{\text{CH}_4}, \quad (3)$$

which includes the long-term O<sub>3</sub> component (23% of IRF<sub>CH<sub>4</sub></sub>).

[11] It should be noted that these formulae are appropriate for aircraft NO<sub>x</sub> 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

**Table 1.** Global IRFs (mW m<sup>-2</sup> yr (Tg NO<sub>2</sub>)<sup>-1</sup>) From This Study, Calculated Using Equations (1)–(3), With Comparable Values From Earlier Work

Vertical extent and season of pulse, and reference	IRF <sub>O<sub>3</sub></sub> /mW m <sup>-2</sup> yr (Tg NO <sub>2</sub> ) <sup>-1</sup>	IRF <sub>CH<sub>4</sub></sub> /mW m <sup>-2</sup> yr (Tg NO <sub>2</sub> ) <sup>-1</sup>	IRF <sub>net</sub> /mW m <sup>-2</sup> yr (Tg NO <sub>2</sub> ) <sup>-1</sup>
Cruise altitudes, July, this study	4.1	-4.9	-1.9
All altitudes, July, (ST04)	5.1	-4.4	-0.26
All altitudes, Annual [Wild <i>et al.</i> , 2001]	7.9	-4.6	+1.8

**Table 2.** Regional Breakdown (%) of Aircraft NO<sub>x</sub> Emissions at Cruise Altitudes and Resulting IRFs

Region <sup>a</sup>	NO <sub>x</sub> emission (%)	IRF <sub>O<sub>3</sub></sub> (%)	IRF <sub>CH<sub>4</sub></sub> (%)	IRF <sub>net</sub> (%)
1. Trans-Pacific(N)	6.9	10.0	10.0	10.0
2. N. America	31.0	26.9	25.5	22.6
3. Trans-Atlantic	9.2	8.8	7.9	6.2
4. Europe	20.8	14.2	15.4	17.8
5. Asia	16.2	14.1	13.4	12.1
6. Trans-Pacific(C/S)	4.1	8.3	11.3	17.6
7. S. America	3.1	5.1	4.9	4.7
8. Africa & Mid-East	5.6	6.8	5.6	3.0
9. Australasia	3.1	6.0	6.1	6.1

<sup>a</sup>Regions (shown in Figure 1a) are arbitrary and have no policy or other significance.

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

[12] Equations (1)–(3), together with the modelled background NO<sub>x</sub> at 250 hPa and the aircraft NO<sub>x</sub> 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<sub>4</sub> 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<sub>4</sub> and long-term O<sub>3</sub> reduction almost universally exceed the positive IRFs associated with the short-term O<sub>3</sub> increase. This means the more sensitive regions show more negative IRFs, i.e., the NO<sub>x</sub> 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<sub>3</sub> 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<sub>x</sub> emissions is important, similar to our results for surface NO<sub>x</sub> [Derwent *et al.*, 2008]. A major influence is the background NO<sub>x</sub> at the emission site. More polluted sites are less sensitive to the addition of more NO<sub>x</sub>; the sensitivities of O<sub>3</sub> production and CH<sub>4</sub> 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<sub>x</sub> 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<sub>x</sub> emissions released at

198 hPa in four latitude bands, and found the O<sub>3</sub> RF varied from 18 mW m<sup>-2</sup> (Tg NO<sub>2</sub>)<sup>-1</sup> in the tropics to 1.5 mW m<sup>-2</sup> (Tg NO<sub>2</sub>)<sup>-1</sup> 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<sub>2</sub>. If we take the NO<sub>x</sub> to CO<sub>2</sub> emission ratio from the NASA 1992 inventory [Penner *et al.*, 1999], then the IRF for CO<sub>2</sub> is 28 mW m<sup>-2</sup> yr for the 1 Tg NO<sub>2</sub> pulse. As CO<sub>2</sub> is long-lived and well-mixed, this value does not vary with emission location or with season, unlike the NO<sub>x</sub> IRF. Locally, the magnitude of the climate cooling from July aircraft NO<sub>x</sub> emissions approaches the IRF from CO<sub>2</sub> (see Figure 2c: -22 mW m<sup>-2</sup> yr over the central Pacific). Elsewhere, the positive IRF from CO<sub>2</sub> generally dominates over the negative net IRF from the July NO<sub>x</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>x</sub> 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<sub>x</sub> emissions (or indeed all non-CO<sub>2</sub> effects [e.g., Lee *et al.*, 2009]) therefore conceals considerable spatial variation. The near-cancellation found here between the IRF terms for aviation NO<sub>x</sub> also appears to be model-dependent. Consequently further calculations, across a range of models, of the geographical variations in the non-CO<sub>2</sub> 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

- Dentener, F., D. Stevenson, J. Cofala, R. Mechler, M. Amann, P. Bergamaschi, F. Raes, and R. Derwent (2005), The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990–2030, *Atmos. Chem. Phys.*, **5**, 1731–1755.
- Derwent, R. G., W. J. Collins, C. E. Johnson, and D. S. Stevenson (2001), Transient behaviour of tropospheric ozone precursors in a global 3-D CTM and their indirect greenhouse effects, *Clim. Change*, **49**, 463–487, doi:10.1023/A:1010648913655.
- Derwent, R. G., D. S. Stevenson, R. M. Doherty, W. J. Collins, M. G. Sanderson, and C. E. Johnson (2008), Radiative forcing from surface NO<sub>x</sub> emissions: Spatial and seasonal variations, *Clim. Change*, **88**, 385–401, doi:10.1007/s10584-007-9383-8.
- Forster, P. M. F., K. P. Shine, and N. Stuber (2006), It is premature to include non-CO<sub>2</sub> effects of aviation in emission trading schemes, *Atmos. Environ.*, **40**, 1117–1121, doi:10.1016/j.atmosenv.2005.11.005.

- Forster, P., et al. (2007), Changes in atmospheric constituents and in radiative forcing, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 129–234, Cambridge Univ. Press, New York.
- Grewe, V., and A. Stenke (2008), AirClim: An efficient tool for climate evaluation of aircraft technology, *Atmos. Chem. Phys.*, *8*, 4621–4639.
- Isaksen, I. S. A., C. Zerefos, K. Kourtidis, C. Meleti, S. B. Dalsøren, J. K. Sundet, A. Grini, P. Zanis, and D. Balis (2005), Tropospheric ozone changes at unpolluted and semi-polluted regions induced by stratospheric ozone changes, *J. Geophys. Res.*, *110*, D02302, doi:10.1029/2004JD004618.
- Köhler, M. O., G. Radel, O. Dessens, K. P. Shine, H. L. Rodgers, O. Wild, and J. A. Pyle (2008), Impact of perturbations to nitrogen oxide emissions from global aviation, *J. Geophys. Res.*, *113*, D11305, doi:10.1029/2007JD009140.
- Lee, D. S., D. W. Fahey, P. M. Forster, P. J. Newton, R. C. N. Wit, L. L. Lim, B. Owen, and R. Sausen (2009), Aviation and global climate change in the 21st century, *Atmos. Environ.*, *43*, 3520–3537, doi:10.1016/j.atmosenv.2009.04.024.
- Lin, X., M. Trainer, and S. C. Liu (1988), On the nonlinearity of the tropospheric ozone production, *J. Geophys. Res.*, *93*, 15,879–15,888, doi:10.1029/JD093iD12p15879.
- Penner, J. E., D. H. Lister, D. J. Griggs, D. J. Dokken, and M. McFarland (1999), *IPCC Special Report on Aviation and the Global Atmosphere*, Cambridge Univ. Press, New York.
- Schimel, D., et al. (1996), Radiative forcing of climate change, in *Climate Change 1995: The Science of Climate Change*, edited by J. T. Houghton et al., pp. 65–132, Cambridge Univ. Press, New York.
- Schumann, U. (2005), Formation, properties and climatic effects of contrails, *C. R. Phys.*, *6*, 549–565, doi:10.1016/j.crhy.2005.05.002.
- Shindell, D. T., and G. Faluvegi (2009), Climate response to regional radiative forcing during the 20th century, *Nat. Geosci.*, *2*, 294–300, doi:10.1038/ngeo473.
- Stevenson, D. S., R. M. Doherty, M. G. Sanderson, W. J. Collins, C. E. Johnson, and R. G. Derwent (2004), Radiative forcing from aircraft NO<sub>x</sub> emissions: Mechanisms and seasonal dependence, *J. Geophys. Res.*, *109*, D17307, doi:10.1029/2004JD004759.
- Stevenson, D. S., et al. (2006), Multi-model ensemble simulations of present-day and near-future tropospheric ozone, *J. Geophys. Res.*, *111*, D08301, doi:10.1029/2005JD006338.
- Stordal, F., G. Myhre, E. J. G. Stordal, W. B. Rossow, D. S. Lee, D. W. Arlander, and T. Svendby (2005), Is there a trend in cirrus cloud due to aircraft traffic?, *Atmos. Chem. Phys.*, *5*, 2155–2162.
- Wild, O., M. J. Prather, and H. Akimoto (2001), Indirect long-term global radiative cooling from NO<sub>x</sub> emissions, *Geophys. Res. Lett.*, *28*, 1719–1722, doi:10.1029/2000GL012573.

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