
**Comparison of model estimates of the effects of aviation emissions on atmospheric ozone and methane**

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Comparison of model estimates of the effects of aviation emissions on atmospheric ozone and methane

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[1] One of the significant uncertainties in understanding the effects of aviation on climate is the effects of aviation emissions on ozone and atmospheric chemistry. In this study the effects of aviation emissions on atmospheric ozone for 2006 and two projections for 2050 are compared among seven models. The models range in complexity from a two-dimensional coupled model to three-dimensional offline and fully coupled three-dimensional chemistry-climate models. This study is the first step in a critical assessment and comparison among these model results. Changes in tropospheric O3 burdens range from 2.3 Tg-O3/Tg-N to 3.0 Tg-O3/Tg-N, and methane radiative forcings range from −8.3 to −12.5 mW/m² for the 2006 aviation emissions. As a group, the chemistry transport models tend to have similar responses while the fully coupled models tend to separate from this group and do not show similar responses to each other. Citation: Olsen, S. C., et al. (2013), Comparison of model estimates of the effects of aviation emissions on atmospheric ozone and methane, Geophys. Res. Lett., 40, 6004–6009, doi:10.1002/2013GL057660.

1. Introduction

[2] Aviation is an important component of the world economy and demand for aviation and its emissions are expected to increase in the future [Intergovernmental Panel on Climate Change (IPCC), 1999; Macintosh and Wallace, 2009]. While the combustion products from aviation (mainly carbon dioxide (CO₂), water vapor (H₂O), nitrogen oxides (NOₓ= NO + NO₂), VOCs (volatile organic compounds), sulfur oxides, soot, and other aerosol components) are similar to those from other forms of transportation, they are unique since they are emitted predominantly at aircraft cruise altitudes from 8 to 12 km. In this upper troposphere-lower stratosphere (UTLS) region, the ozone production efficiency of NOₓ emissions is much greater than at the surface [IPCC, 1999; Gauss et al., 2006; Köhler et al., 2008] and the radiative impact of changes in ozone is also greater than at the surface [e.g., Lacis et al., 1990; Forster and Shine, 1997]. There have been many studies and intercomparisons of the effects of aviation on atmospheric chemistry [e.g., Brasseur et al., 1998; IPCC, 1999, Grewe et al., 2002; Köhler et al., 2008; Hoor et al., 2009; Hodnebrog et al., 2011; Mohre et al., 2011; Köhler et al., 2013; Jacobson et al., 2013]. This study examines the effects of aviation emissions on atmospheric O₃ and CH₄ among recent versions of seven established atmospheric chemistry models that have just recently been used to evaluate aviation effects.

[3] Aviation emissions lead to an increase in ozone and the hydroxyl radical (OH), the main oxidant in the troposphere. The increased OH concentrations in turn lead to an increase in CH₄ chemical destruction and a decrease in CH₄ concentration. Since ozone and methane are both greenhouse gases, the changes in global average radiative forcing (RF) due to the increase in ozone and decrease in methane offset each other to some degree although the magnitude of the cancellation is quite uncertain [e.g., IPCC, 1999, Fuglestvedt et al., 1999; Stevenson et al., 2004; Köhler et al., 2008; Hoor et al., 2009; Hodnebrog et al., 2011; Holmes et al., 2011; Myhre et al., 2011].

[4] The largest effects on atmospheric ozone from aviation are due to aviation NOₓ emissions with the largest impact occurring in the main flight corridors in the midlatitude Northern Hemisphere at cruise altitude [Brasseur et al., 1998; IPCC, 1999, Grewe et al., 2002; Köhler et al., 2008; Hoor et al., 2009; Stevenson et al., 2009; Hodnebrog et al., 2011; Myhre et al., 2011; Köhler et al., 2013]. It has been estimated that aviation NOₓ emissions increase tropospheric ozone by 5.5 to 16.4 Tg-O3/Tg-N [e.g., Lee et al., 2010]. A multimodel comparison estimates the tropospheric O₃ burden to be 344 ± 39 Tg [Stevenson et al., 2006]. Ozone is a relatively short-lived gas in the UTLS region, so the perturbation and associated RF are largely limited to the hemisphere where the NOₓ emissions are released while CH₄ is a long-lived gas with a lifetime of around a decade, and thus, its changes tend to affect the entire atmosphere. The decrease in background CH₄ tends to cause a decrease in background tropospheric
ozone, e.g., the so-called long-lived O$_3$ effect making a negative contribution to RF [Prather, 1994; Wild and Prather, 2000; Hoor et al., 2009; Hodnebrog et al., 2011; Myhre et al., 2011]. A recent review of uncertainties in aviation O$_3$-CH$_4$ radiative forcing estimates reports a short-lived O$_3$ RF due to aviation of $27.3 \pm 9.7$ mW/m$^2$ and CH$_4$ RF of $-16.1 \pm 5.6$ mW/m$^2$ scaled to a 1 Tg-N/yr emission [Holmes et al., 2011]. Due to the long CH$_4$ lifetime, most (but not all) three-dimensional model simulations of atmospheric chemistry, including those to evaluate the effects of aviation, prescribe atmospheric CH$_4$ concentrations in the lower model levels instead of using a CH$_4$ surface flux. Estimates of changes in CH$_4$ and long-lived O$_3$ are then estimated from relationships between changes in the CH$_4$ lifetime between simulations with and without aviation emissions [e.g., Fuglestvedt et al., 1999] (Section S3 in the supporting information).

Aviation effects are typically evaluated from the difference between two simulations, one with aviation emissions and one without. We use this approach which is suitable when the response is generally linear with the emissions; however, other methods are also in use, e.g., emissions scaling [Hoor et al., 2009; Grewe et al., 2010; Hodnebrog et al., 2011; Myhre et al., 2011].

The Federal Aviation Administration’s (FAA) Aviation Climate Change Research Initiative (ACCRI) supports research into the effects of aviation on atmospheric chemistry and climate. This paper compares simulations among seven models participating in ACCRI to evaluate the effects of current and future aviation emissions on atmospheric O$_3$ and CH$_4$. Some of these models have not previously been used to evaluate the effects of aviation while others are updated versions of previously used models. Here we compare and contrast the simulated perturbations due to aviation emissions. We also investigate commonalities and differences in the model responses relative to their complexity. In a detailed comparison of the model simulated background atmospheres relative to observations and ozone sensitivity to NO$_x$ perturbations, (G. Brasseur et al., Model Intercomparison of Ozone Sensitivity to NO$_x$ emissions in the vicinity of the extratropical tropopause, submitted to Geophysical Research Letters, 2013) noted some striking differences between the models. The simulations utilize recent estimates of aviation emissions for 2006 and two scenarios for 2050 derived from the Aviation Environmental Design Tool (AEDT).

2. Data and Models

2.1. Emissions

The aviation emissions used in this study are derived from the Aviation Environmental Design Tool (AEDT) [Roof et al., 2007; Barrett et al., 2010]. These data sets include estimates of aviation fuelburn and emissions of NO$_x$, VOCs, carbon monoxide, carbon dioxide, water vapor, and aerosols for 2006 [Wilkinson et al., 2010; Olsen et al., 2012] as well as two projections for 2050. The 2006 emissions have been compared against other aviation emissions data sets [Wilkinson et al., 2010; Olsen et al., 2012]. The first 2050 scenario (Base) assumes increases in aviation emissions are directly related to projected increases in demand with no technology improvements. The second scenario (Scen1) assumes the same increase in demand but also assumes technological and operational improvements in efficiency. Annual NO$_x$ emissions for the 2006 scenario are 0.8 Tg-N/yr and emissions for the 2050 Base and Scen1 scenarios are 4.0 Tg-N/yr and 1.6 Tg-N/yr, respectively (Section S1).

For all of the model simulations, background emissions of nonaviation shorter-lived species (e.g., NO$_x$ and VOCs) and prescribed concentrations in the lower model levels for longer-lived species, e.g., CH$_4$, chlorofluorocarbons (CFCs), and nitrous oxide (N$_2$O), are from the Intergovernmental Panel on Climate Change (IPCC) Representative Concentration Pathway (RCP) 4.5 scenario [Thomson et al., 2011; van Vuuren et al., 2011] for the appropriate year, except as noted. The IPCC RCP 4.5 scenario was chosen since it represents a midrange future growth path.

2.2. Model Descriptions

The models included in this study encompass a wide range of detail and complexity. Three of the models are three-dimensional (3-D) chemistry transport models (CTMs) driven off-line meteorology: Goddard Earth Observing System Chemistry (GEOS-Chem, http://geos-chem.org) and the Community Atmosphere Model versions 4 [Lamarque et al., 2012] and 5 [Liu et al., 2012] (CAM4 and CAM5) which are the atmospheric component models of the Community Earth System Model. One is a chemistry-climate model (CCM) without aerosol-climate coupling (the Goddard Earth Observing System Chemistry-Climate Model (GEOS CCM) version 3) [Oman et al., 2011] and two are CCMs with aerosol-climate coupling (the Gas, Aerosol, Transport, Radiation, General Circulation, Mesoscale, Ocean Model (GATOR-GCMOM) [Jacobson et al., 2011, 2013] and the National Aeronautics and Space Administration (NASA) ModelE2 [Shindell et al. 2006]). One (GATOR-GCMOM) treats aircraft exhaust from each flight worldwide at the subgrid scale. Finally, the Integrated Global System Model (IGSM) is a two-dimensional (2-D) Earth system model of intermediate complexity [Sokolov et al., 2005]. While the IGSM model does not have as complete a representation of atmospheric chemistry and physics as the other models, this type of intermediate complexity model is useful for policy analyses due to its relatively low computational requirements, and thus, it is important to examine its performance relative to the 3-D models. Detailed descriptions of the models and O$_3$ and CH$_4$ radiative forcing calculations are provided in the supporting information (Section S2 and Table S2).

3. Results and Discussion

3.1. Concentration Profiles

In all of the models the effect of aviation emissions on O$_3$ is mostly positive in the Northern Hemisphere middle to upper troposphere with a relatively small effect in the Southern Hemisphere (Figure S1). There are, however, substantial differences with respect to the magnitude of the effect on ozone with the NASA ModelE2 having a lower response than the other models. The largest effects occur in the region from 30°N to 60°N, so most of our analyses will focus on this region. In all of the models except one, the peak absolute increase occurs around 10 to 12 km and ranges from about 5 to 8 ppb (Figure 1). The GATOR-GCMOM O$_3$ peak occurs at a slightly higher altitude than the other 3-D models. It also maintains the largest vertical gradient between cruise altitudes and the lower troposphere with the aviation O$_3$ perturbation being relatively localized around cruise altitudes which may be due to its higher vertical resolution in this region (~0.5 km
versus ~1 km for most of the other models). The other models’ perturbations are less localized and suggest more downward vertical mixing, perhaps indicative of diffusion or differences in convective mixing. It is notable that for the NASA ModelE2, there is a decrease in O₃ above ~10 km which is largest at 12 km. There is also a large decrease in O₃ for GATOR-GCMOM above ~14 km which reaches nearly 35 ppb by 16 km; this decrease is caused by aviation-induced upper-tropospheric increases in stability that reduce the transport of ozone vertically to above the aircraft layer [Jacobson et al., 2013]. Finally, there is a decrease in O₃ above 11 km in GEOS-CCM, but at 60–90°N and 40–90°S (Figure S1). For the 3-D models, the peaks in relative O₃ changes typically occur about 1 to 2 km lower than the peak absolute increases (Figure 1) consistent with other models [e.g., Lee et al., 2010] and range from about 4 to 6%. IGSM has the largest relative increase (9%) and it occurs at the same altitude as the absolute peak.

Similar to the O₃ and NOₓ aviation perturbations, the NOᵧ aviation perturbation is largest at cruise altitudes (Figure 1). Although the content of NOᵧ (=NOₓ + HNO₃ + 2*N₂O₅ + PAN + Nitrate aerosols) varies somewhat by model, the major species are present in all models. The perturbations range from 0.07 ppb (IGSM) to 0.5 ppb (NASA ModelE2) or ~20% for the NASA ModelE2 up to about 50% for GEOS-Chem. In IGSM there is a large increase in the relative perturbation above 12 km.

The HOₓ (=OH + HO₂) perturbations due to aviation emissions are generally small except near cruise altitudes for the 3-D models (Figure 1). For all of the models except one, HOₓ decreases at cruise altitudes. Since aviation emissions generally increase OH, this indicates enhanced HO₂ loss. GEOS-Chem shows the largest decrease of 0.9 ppt. The other models’ decreases range from 0.2 to 0.5 ppt (parts per trillion). In the GATOR-GCMOM simulations cruise altitude HOₓ increases by ~0.5 ppt. This may be due to lower NOₓ in GATOR-GCMOM due to conversion to nitrate aerosol, which affects HOₓ chemistry and the inclusion of aviation H₂O emissions; however, other models which also included aviation H₂O emissions do not show an increase in HO₂. While IGSM also shows a decrease in HOₓ at cruise altitudes, it also shows a decrease at lower altitudes.

### 3.2. Tropospheric Burdens and Changes

In the following analyses, the troposphere is defined as the region where O₃ concentrations are less than 150 ppb in the baseline nonaviation simulations [e.g., Prather et al., 2001, Stevenson et al., 2006]. Global tropospheric ozone burdens for the baseline simulations range from about 275 Tg for IGSM to 380 Tg for the CAM4 simulations (Table 1). These are within the range of model results reported in Stevenson [2000].
et al. [2006] but not all are within the reported standard deviation. Changes in the tropospheric ozone burden due to aviation emissions range from 2.3 to 9.1 Tg-O_3 for the AEDT 2006 aviation NO_x emissions (0.8 Tg-N) (Table 1). These changes generally fall into two groups with CAM4, CAM5, GEOS-C CM, and GEOS-Chem having higher responses and NASA ModelE2, GATOR-GCMOM, and IGSM having lower responses. This grouping follows through to the 2050 Scenarios where the CAM4 response is considerably higher than either IGSM or NASA ModelE2, e.g., 29 Tg-O_3 (CAM4) versus 13.7 Tg-O_3 (IGSM) and 10.3 Tg-O_3 (NASA ModelE2) for the 2050 Base Scenario (Table 1). For the 2006 emissions the relative changes range from a 0.7% (NASA ModelE2) to 2.5% (GEOS CCM and GEOS-Chem).

[15] Normalized changes in tropospheric ozone burden due to aviation emissions range from 2.5 to 11 Tg-O_3 per Tg-N of aviation emissions (Figure 2). These changes correspond to tropospheric ozone burden increases of increases of 0.9 to 3.1%/Tg-N. In contrast to some of the profile changes, the groupings between higher and lower response models tend to be the same for absolute as well as relative O_3 burden changes. It is notable that some of the values reported here are outside of the 5–16 Tg-O_3/Tg-N range reported in Lee et al. [2010]. There is some nonlinearity in the response at large aviation NO_x emissions in the models that reported the AEDT 2050 simulations. The decrease in dO_3/Tg-N is less pronounced in the NASA ModelE2 than in the other models (Figure 2). It is worth noting that the nonlinearity over this range of aviation NO_x emissions is much smaller than the spread between the models.

### 3.3. O_3 and CH_4 Radiative Forcing

Five models reported radiative forcings. The differences in O_3 changes due to aviation lead to a relatively large range in the ozone radiative forcings. The O_3-short instantaneous RFs for the AEDT 2006 emissions range from 6.4 mW/m² for NASA ModelE2 to 36.5 mW/m² for CAM4 (Table 2). For the 2050 Base and Scen1 scenarios, the highest RFs are 143.0 mW/m² and 70.4 mW/m² (CAM4), and the lowest are 28.5 mW/m² and 13.4 mW/m² (NASA ModelE2). The radiative forcing normalized by aviation NO_x emissions ranges from a high of 45 mW/m²/Tg-N for CAM4 with the AEDT 2006 emissions to 7 mW/m²/Tg-N for NASA ModelE2 with the AEDT 2050 Base emissions. These values (except for NASA ModelE2) are substantially higher than the range reported by Myhre et al. [2011] of 15–25 mW/m²/Tg-N from an intercomparison of five models. The CAM4 and GEOS CCM are above the mean ± standard deviation estimate of 17.6 to 37 mW/m²/Tg-N reported in the Holmes et al. [2011] review but within the reported range of the models (−15 to 45 mW/m²/Tg-N), while the NASA ModelE2 O_3 RF is outside of this range. The CH_4 RFs reported here do not include changes in stratospheric water vapor due to changes in CH_4 and are not adjusted for the history of emissions. For the AEDT 2006

<table>
<thead>
<tr>
<th>Model</th>
<th>Background</th>
<th>Aviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>CAM4</td>
<td>373</td>
<td>7.3 (2.0)</td>
</tr>
<tr>
<td>IGSM</td>
<td>275</td>
<td>4.5 (1.6)</td>
</tr>
<tr>
<td>NASA ModelE2</td>
<td>350</td>
<td>2.3 (0.7)</td>
</tr>
<tr>
<td>CAM5</td>
<td>318</td>
<td>5.4 (1.7)</td>
</tr>
<tr>
<td>GEOS CCM</td>
<td>327</td>
<td>6.0 (1.8)</td>
</tr>
<tr>
<td>GEOS-Chem</td>
<td>363</td>
<td>9.1 (2.5)</td>
</tr>
<tr>
<td>GATOR-GCMOM</td>
<td>280</td>
<td>2.5 (2.3)</td>
</tr>
</tbody>
</table>

Table 1. Tropospheric Ozone Burdens for the Background Atmosphere (Tg-O_3) and Changes Due to Aviation Emissions (Tg-O_3 (%))

<table>
<thead>
<tr>
<th>Model</th>
<th>Background</th>
<th>Aviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>CAM4</td>
<td>369</td>
<td>28.6 (7.8)</td>
</tr>
<tr>
<td>IGSM</td>
<td>281</td>
<td>15.7 (4.9)</td>
</tr>
<tr>
<td>NASA ModelE2</td>
<td>330</td>
<td>10.3 (3.1)</td>
</tr>
<tr>
<td>CAM5</td>
<td>318</td>
<td>23.4 (7.3)</td>
</tr>
<tr>
<td>GEOS CCM</td>
<td>318</td>
<td>27.0 (8.5)</td>
</tr>
<tr>
<td>GEOS-Chem</td>
<td>318</td>
<td>11.0 (3.5)</td>
</tr>
<tr>
<td>GATOR-GCMOM</td>
<td>280</td>
<td>2.5 (2.3)</td>
</tr>
</tbody>
</table>

Table 2. Global Average Aviation-Induced O_3 Short and CH_4 Radiative Forcing (mW/m² (mW/m²/Tg-N))

<table>
<thead>
<tr>
<th>Model</th>
<th>O_3</th>
<th>CH_4</th>
</tr>
</thead>
<tbody>
<tr>
<td>CAM4</td>
<td>36.5 (45)</td>
<td>−12.5 (−15)</td>
</tr>
<tr>
<td>CAM5</td>
<td>24.5 (30)</td>
<td>−11.2 (−13)</td>
</tr>
<tr>
<td>NASA ModelE2</td>
<td>6.4 (8)</td>
<td>−8.3 (−10)</td>
</tr>
<tr>
<td>IGSM</td>
<td>26.0 (32)</td>
<td>−9.7 (−12)</td>
</tr>
<tr>
<td>GEOS CCM</td>
<td>30.5 (38)</td>
<td>−12.3 (−15)</td>
</tr>
<tr>
<td>GATOR-GCMOM</td>
<td>143.0 (36)</td>
<td>70.6 (−18)</td>
</tr>
<tr>
<td>IGSM</td>
<td>22.5 (19)</td>
<td>−55.3 (−14)</td>
</tr>
<tr>
<td>NASA ModelE2</td>
<td>28.5 (7)</td>
<td>−41.0 (−10)</td>
</tr>
<tr>
<td>CAM4</td>
<td>143.0 (36)</td>
<td>50.6 (−13)</td>
</tr>
<tr>
<td>CAM5</td>
<td>111.0 (28)</td>
<td>−72.1 (−18)</td>
</tr>
<tr>
<td>NASA ModelE2</td>
<td>28.5 (7)</td>
<td>−31.0 (−20)</td>
</tr>
<tr>
<td>IGSM</td>
<td>80.0 (20)</td>
<td>52.7 (−16)</td>
</tr>
<tr>
<td>GEOS CCM</td>
<td>162.3 (41)</td>
<td>−30.5 (−23)</td>
</tr>
</tbody>
</table>

*GEOES-Chem and GATOR-GCMOM did not report radiative forcings.
emissions, there is less of a spread between CH\textsubscript{4} RFs which range from \(-8.3\) mW/m\(^2\) for the NASA ModelE2 to \(-12.5\) mW/m\(^2\) for CAM4. All of the models fall within the mean \pm standard deviation estimate of \(-10.5\) to \(-21.7\) W/m\(^2\)
Tg-N reported in Holmes et al. [2011]. For all of the models except NASA ModelE2, the sum of the O\textsubscript{3} short-term and CH\textsubscript{4} RFs are positive. For all of the NASA ModelE2 simulations, they are negative (Table 2). Previous published results suggest that the net forcing from O\textsubscript{3} short and CH\textsubscript{4} is positive [e.g., IPCC, 1999; Lee et al., 2010; Hodnebrog et al., 2011; Holmes et al., 2011; Myhre et al., 2011].

4. Conclusions

[17] This paper compares simulations among seven models to evaluate the effects of current and future aviation emissions on atmospheric O\textsubscript{3} and CH\textsubscript{4}. The seven models show a large range in the simulated changes due to aviation emissions. This is likely due to differences in the details of their representations of the physics and chemistry of the background atmosphere. This study is the beginning phase of a full evaluation of these differences and their effects on climate. The offline model results as a group (e.g., CAM4, CAM5, and GEOS-Chem) tend to be more similar in their response and sensitivities. While one might expect the 3-D fully coupled models (GEOS CCM, GATOR-GCMOM, and NASA ModelE2) to perform similarly (but differently from the offline models) due to the inclusion of more feedback processes and coupled interactions (particularly aerosol and cloud coupling processes), this is not the case for the models examined here. The fully coupled models, where they are different from the offline models, often respond quite differently from each other, e.g., for NO\textsubscript{x} and O\textsubscript{3}. These differences likely result from differences in the details of the implementation of aerosol coupling processes, model resolution, and treatments of physical and numerical diffusion. Although IGS is a zonal mean model with a reduced tropospheric chemical scheme, it is generally performed within the envelope of the 3-D models. The range of these results suggests that there remain uncertainties in quantifying the effect of aviation emissions on ozone and point to the necessity of more detailed critical testing of models versus observations as perhaps the most important path to reducing these uncertainties.

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