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 O_3 burdens range from 2.3 Tg-O_3/Tg-N to 3.0 Tg-O_3/Tg-N, ozone radiative forcings range from 6 to 37 mW/m^2, and methane radiative forcings range from −8.3 to −12.5 mW/m^2 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 (CO_2), water vapor (H_2O), nitrogen oxides ($NO_x = NO + NO_2$), 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 NO_x 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; *Myhre et al*., 2011; *Köhler et al*., 2013; *Jacobson et al*., 2013]. This study examines the effect of aviation emissions on atmospheric O_3 and CH_4 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 CH_4 chemical destruction and a decrease in CH_4 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 cancelation 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 NO_x 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 NO_x emissions increase tropospheric ozone by 5.5 to 16.4 Tg-O_3/Tg-N [e.g., *Lee et al*., 2010]. A multimodel comparison estimates the tropospheric O_3 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 NO_x emissions are released while CH_4 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 CH_4 tends to cause a decrease in background tropospheric

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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 ± 9.7 mW/m^2 and CH_4 RF of -16.1 ± 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).

[5] 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].

[6] 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

[7] 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 [*Wilkerson 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 [*Wilkerson 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).

[8] 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_2O), 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

[9] 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 with offline 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

[10] 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

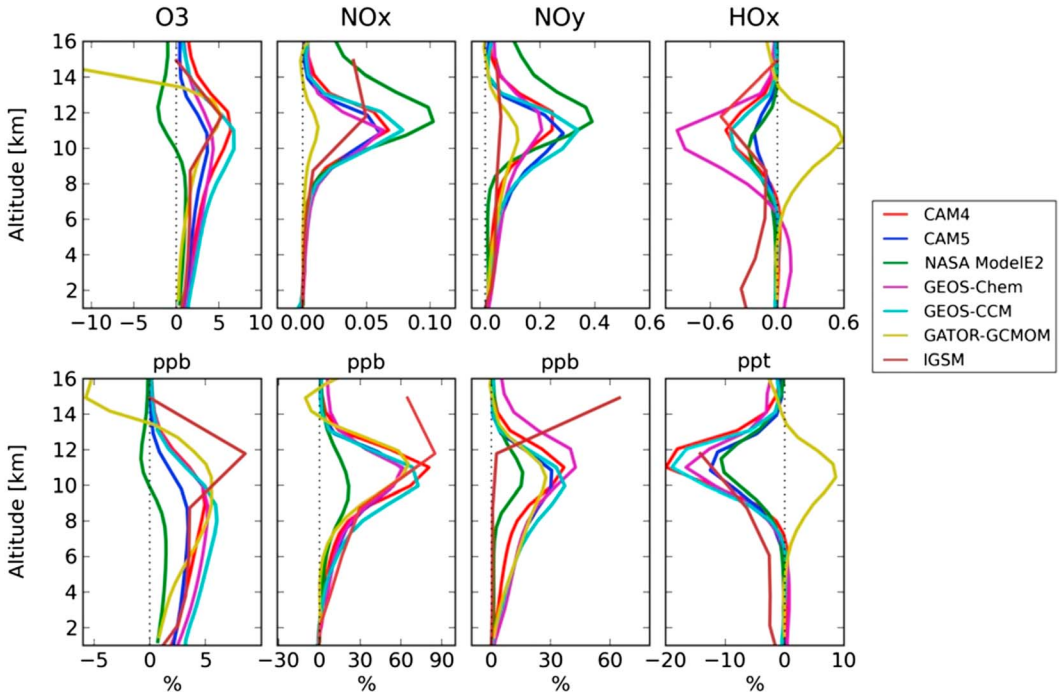

Figure 1. Effect of aviation emissions on O_3, NO_x, NO_y, and HO_x. Profiles are zonal mean averaged over 30°N to 60°N. (top row) Absolute perturbation and (bottom row) percent perturbation. Perturbations are for the AEDT 2006 aviation emissions. CAM4, CAM5, and GEOS-Chem are CTMs, NASA ModelE2, GEOS CCM, and GATOR-GCMOM are CCMs, and IGSM is a 2-D model of intermediate complexity.

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 O_3 above ~ 10 km which is largest at 12 km. There is also a large decrease in O_3 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 O_3 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 O_3 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.

[11] The aviation NO_x perturbations are largest at cruise altitudes. The NASA ModelE2 has the largest perturbation of around 0.11 ppb, and GATOR-GCM has the lowest at ~ 0.014 ppb with the other models grouped around 0.07 ppb. The relative perturbations also peak at cruise altitudes and are mostly similar (~80%) except for the NASA ModelE2 which is ~20%. Although the GATOR-GCMOM NO_x absolute perturbation is smaller than the other models', the relative change is close to that of the other models due its lower background NO_x concentrations from the conversion of NO_x to nitrate aerosol and dissolution of NO_x based on solubility [*Brasseur et al*., this issue]. The high background NO_x concentrations in the NASA ModelE2 [*Brasseur et al*., this issue] lead to smaller relative changes even though it has the largest absolute NO_x perturbation.

[12] Similar to the O_3 and NO_x aviation perturbations, the NO_y aviation perturbation is largest at cruise altitudes (Figure 1). Although the content of NO_y (=NO_x + HNO_3 + 2*N_2O_5 + 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.

[13] The HO_x (=OH + HO_2) 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, HO_x decreases at cruise altitudes. Since aviation emissions generally increase OH, this indicates enhanced HO_2 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 HO_x increases by ~ 0.5 ppt. This may be due to lower NO_x in GATOR-GCMOM due to conversion to nitrate aerosol, which affects HO_x chemistry and the inclusion of aviation H_2O emissions; however, other models which also included aviation H_2O emissions do not show an increase in HO_2. While IGSM also shows a decrease in HO_x at cruise altitudes, it also shows a decrease at lower altitudes.

3.2. Tropospheric Burdens and Changes

[14] In the following analyses, the troposphere is defined as the region where O_3 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*

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

	2006		2050		
Model[a]	Background	Aviation	Background	Base	Scen1
CAM4	373	7.3 (2.0)	369	28.6 (7.8)	13.8 (3.7)
IGSM	275	4.5 (1.6)	281	13.7 (4.9)	6.2 (2.2)
NASA ModelE2	350	2.3 (0.7)	330	10.3 (3.1)	4.5 (1.4)
CAM5	318	5.4 (1.7)	318	23.4 (7.3)	11.0 (3.5)
GEOS CCM	327	6.0 (1.8)	318	27.0 (8.5)	11.0 (3.5)
GEOS-Chem	363	9.1 (2.5)			
GATOR-GCMOM	280	2.5 (2.3)			

[a]CAM4, CAM5, and GEOS-Chem are CTMs; NASA ModelE2, GEOS CCM, and GATOR-GCMOM are CCMs; and IGSM is a 2-D model of intermediate complexity. The troposphere is defined as the region with $O_3 < 150$ ppb. GEOS-Chem and GATOR-GCMOM did not report results for the 2050 scenarios.

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-CCM, 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.

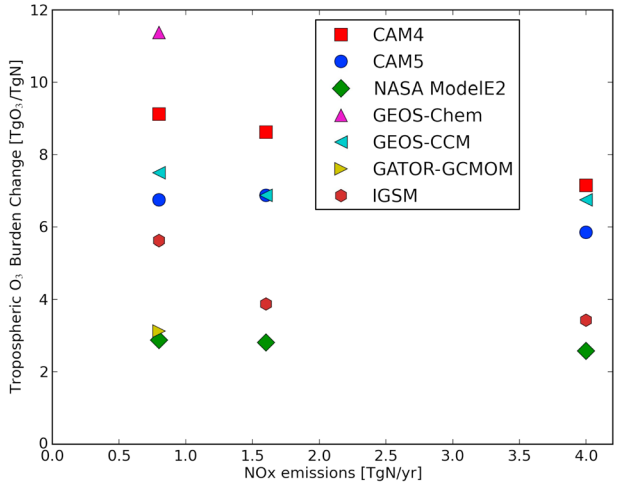

Figure 2. Normalized tropospheric ozone burden changes due to aviation emissions for the AEDT 2006 emissions and AEDT 2050 Base and Scen1 emissions, units are Tg-O_3 per Tg-N emitted by aviation.

3.3. O_3 and CH_4 Radiative Forcing

[16] 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/m^2 for NASA ModelE2 to 36.5 mW/m^2 for CAM4 (Table 2). For the 2050 Base and Scen1 scenarios, the highest RFs are 143.0 mW/m^2 and 70.4 mW/m^2 (CAM4), and the lowest are 28.5 mW/m^2 and 13.4 mW/m^2 (NASA ModelE2). The radiative forcing normalized by aviation NO_x emissions ranges from a high of 45 mW/m^2/Tg-N for CAM4 with the AEDT 2006 emissions to 7 mW/m^2/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/m^2/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/m^2/Tg-N reported in the *Holmes et al*. [2011] review but within the reported range of the models (~15 to 45 mW/m^2/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 2. Global Average Aviation-Induced O_3 Short and CH_4 Radiative Forcing (mW/m^2 (mW/m^2/Tg-N))[a]

	O_3	CH_4
	2006	
CAM4	36.5 (45)	−12.5 (−15)
CAM5	24.5 (30)	−11.2 (−13)
NASA ModelE2	6.4 (8)	−8.3 (−10)
IGSM	26.0 (32)	−9.7 (−12)
GEOS CCM	30.5 (38)	−12.3 (−15)
	2050 Base	
CAM4	143.0 (36)	−70.6 (−18)
CAM5	111.0 (28)	−55.3 (−14)
NASA ModelE2	28.5 (7)	−41.0 (−10)
IGSM	80.0 (20)	−50.6 (−13)
GEOS CCM	162.3 (41)	−72.1 (18)
	2050 Scen1	
CAM4	70.4 (45)	−31.0 (−20)
CAM5	52.4 (33)	−25.7 (−16)
NASA ModelE2	13.4 (9)	−17.0 (−11)
IGSM	37.0 (24)	−27.7 (−18)
GEOS CCM	67.2 (40)	−35.5 (−23)

[a]GEOS-Chem and GATOR-GCMOM did not report radiative forcings.

emissions, there is less of a spread between CH_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 ± 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_3 short-term and CH_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_3 short and CH_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_3 and CH_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_x and O_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 IGSM 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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