



# **Journal of Geophysical Research: Atmospheres**

# **RESEARCH ARTICLE**

10.1002/2016JD025594

#### **Kev Points:**

- All-altitude aircraft emissions in five models estimate an increase in 2006 surface-layer ozone of 0.3 to 1.9% and PM<sub>2.5</sub> by -1.9 to 1.2%
- Modeled O<sub>3</sub> and PM<sub>2.5</sub> response to aircraft is greatly affected by the inclusion of feedbacks between aircraft emissions and meteorology
- While some models show much of the surface-layer perturbations stem from emissions >1 km, they are minor compared with perturbations aloft

#### **Supporting Information:**

- Data Set S1
- Data Set S2
- · Supporting Information S1

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#### Citation:

Cameron, M. A., et al. (2017), An intercomparative study of the effects of aircraft emissions on surface air quality, *J. Geophys. Res. Atmos.*, 122, 8325–8344, doi:10.1002/2016JD025594.

Received 28 JUN 2016 Accepted 19 JUL 2017 Accepted article online 24 JUL 2017 Published online 5 AUG 2017

# An intercomparative study of the effects of aircraft emissions on surface air quality

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**Abstract** This study intercompares, among five global models, the potential impacts of all commercial aircraft emissions worldwide on surface ozone and particulate matter (PM<sub>2.5</sub>). The models include climate-response models (CRMs) with interactive meteorology, chemical-transport models (CTMs) with prescribed meteorology, and models that integrate aspects of both. Model inputs are harmonized in an effort to achieve a consensus about the state of understanding of impacts of 2006 commercial aviation emissions. Models find that aircraft increase near-surface ozone (0.3 to 1.9% globally), with qualitatively similar spatial distributions, highest in the Northern Hemisphere. Annual changes in surface-level PM<sub>2.5</sub> in the CTMs (0.14 to 0.4%) and CRMs (–1.9 to 1.2%) depend on differences in nonaircraft baseline aerosol fields among models and the inclusion of feedbacks between aircraft emissions and changes in meteorology. The CTMs tend to result in an increase in surface PM<sub>2.5</sub> primarily over high-traffic regions in the North American midlatitudes. The CRMs, on the other hand, demonstrate the effects of aviation emissions on changing meteorological fields that result in large perturbations over regions where natural emissions (e.g., soil dust and sea spray) occur. The changes in ozone and PM<sub>2.5</sub> found here may be used to contextualize previous estimates of impacts of aircraft emissions on human health.

Plain Language Summary This study uses five global atmospheric computer models to estimate the effects of global aircraft emissions on surface air quality by tracking changes in ozone and small particles. While each model uses different modeling techniques, they are harmonized with identical input in order to resolve the somewhat conflicting results of previous studies. The results indicate that all-altitude 2006 commercial aircraft increase global, annual surface ozone by 0.3 to 1.9% and small particulate matter by –1.9 to 1.2%. While the models show general agreement in the ozone response, the response to small particles varies significantly depending on whether models simulate two-way chemistry/meteorology "feedbacks" (i.e., meteorology affects emissions and emissions affect meteorology) or one-way (meteorology affects emissions, but meteorology remains the same in simulations with and without aircraft). In models with feedbacks, most of the change to surface particles comes from changes to natural background particles (from sea spray and soil dust), and not aviation directly. This study helps explain the reasons for disagreement in previous studies and emphasizes the need to further investigate the effects of feedbacks in atmospheric models.

#### 1. Introduction

As the popularity of and global access to air travel expands, quantifying its impact on air pollution becomes increasingly important. However, modeling these effects remains difficult as it requires representing spatial scales of both the global distribution and spread of aircraft emissions, as well as the nonlinear microphysical and chemical processes within an individual plume. In addition, the majority of aircraft emissions are released well above the atmospheric boundary layer, so models require accurate chemical, microphysical, and meteorological representations in order to capture relevant changes in surface air quality. These same spatial and temporal challenges make it virtually impossible for air quality measurement campaigns to disentangle the

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effects of aircraft emissions from those of other sources. As such, computer models simulating the global atmosphere currently provide the best estimate of the effects of all-altitude aircraft on human health.

Several studies have assessed the effects of aviation on the global atmosphere [e.g., Friedl, 1997; Brasseur et al., 1998; Penner et al., 1999; Lee et al., 2009; Søvde et al., 2014; Brasseur et al., 2016], though surface-level results are only a minor focus. Conversely, studies that do emphasize surface air quality effects primarily investigate the local response using emissions near airports, rather than global, all-altitude emissions [see Masiol and Harrison, 2014, and references therein]. Tarrasón et al. [2004], Barrett et al. [2010], Lee et al. [2013], Jacobson et al. [2013], and Morita et al. [2014] do model the impacts of cruise-altitude emissions on surface air quality and premature mortalities, though each produced a different range of results; inconsistent methodologies, reported metrics, and model characteristics make it difficult to distinguish the causes for the large spread in results. This work builds upon those previous studies by intercomparing five global atmospheric models, all previously used and validated in aircraft emissions studies [e.g., Brasseur et al., 2016; Olsen et al., 2013; Jacobson et al., 2013; Unger et al., 2010; Gettelman and Chen, 2013], to examine the impacts of 2006 all-altitude aircraft emissions on surface air quality in greater detail and pinpoint the major sources of disagreement in previous studies.

Degraded air quality has been shown not only to adversely affect human health [e.g., *Ostro et al.*, 2006; *Brunekreef and Holgate*, 2002; *Jerrett et al.*, 2009; *Ji et al.*, 2011; *Levy et al.*, 2012] but also to decrease atmospheric visibility; cause damage to crops, buildings, and exposed materials; and affect climate. While there are climatic effects of carbon dioxide, methane, water vapor, and other greenhouse gases that influence air quality, this study limits itself primarily to surface (bottom layer) ozone (O<sub>3</sub>) and fine particulate matter (PM<sub>2.5</sub>; less than 2.5 µm in diameter), two of six criteria pollutants regulated by the U.S. Environmental Protection Agency to protect human health and welfare. Ozone, PM<sub>2.5</sub>, and their precursors can be produced from aviation emissions at ground level during landing and takeoff or transported from cruise-level to the surface. Surface-layer ozone is produced in the daytime by chemical reactions involving nitrogen oxides and volatile organic compounds emitted by aircraft. PM<sub>2.5</sub> components are emitted directly by aircraft (black carbon, primary organic matter, and sulfate) or formed based on interactions between emissions and the background atmosphere.

# 1.1. Advancements in Air Quality Modeling

Computational demands restrict the level of integration between climate, chemistry, and aerosol calculations in global air quality models. Accurately modeling ozone and  $PM_{2.5}$  concentrations depends on nonlinear, time-dependent changes in emissions, chemistry, photolysis, transport, clouds, and aerosols, which in turn further affect a number of modeled variables. Capturing these interactions at the appropriate spatial and temporal scales is particularly difficult in the context of aircraft emissions, which are both globally distributed and often deposited as concentrated plumes into otherwise clean air aloft.

Historically, chemical-transport models (CTMs) have been primarily used to model changes in air quality. CTMs use prescribed meteorological fields, obtained off-line, to drive changes in the transport and evolution of chemical and particulate emissions. This one-way interaction, in which meteorology affects changes in emissions but emissions do not affect meteorology, simplifies dynamics and can allow for a more sophisticated representation of chemistry.

In the last few years, however, computational and algorithmic advancements have resulted in a trend for modelers to integrate chemical and aerosol changes into meteorological calculations with varying levels of complexity [Baklanov et al., 2011]. While this coupling of physical, dynamic, and chemical processes has been recognized as a milestone in the development of next-generation, state-of-the-science models [e.g., Grell and Baklanov, 2011; Zhang et al., 2012; Bauer et al., 2015], there is an increased need for studies exploring these complex interactions, as model analysis catches up with model capabilities. Recently, the Air Quality Model Evaluation International Initiative Phase II (AQMEII-2) study compared multiple air quality models capable of running with or without coupling. Among others, this resulted in studies investigating the models' ability to accurately predict background ozone [Im et al., 2015a], and PM [Im et al., 2015b], as well as the effects of aerosol feedbacks on weather [Makar et al., 2015a], and chemistry [Makar et al., 2015b] over North America and Europe. As more models migrate toward fully coupled integration of systems, model evaluations will also need to adapt to include multimodel validations and intercomparisons such as this [Baklanov et al., 2014].



For comparison with models used in the present work, models that fully couple changes in aerosol and chemical composition with online meteorological calculations are labeled "climate-response models" (CRMs). That is, aircraft emissions are affected by meteorology and that meteorology is in turn updated at each time step based in part on the changes from emissions. Conversely, meteorology in chemical-transport models (CTMs) do not dynamically update from changes in emissions. These "CRM" and "CTM" labels are added to the following descriptions of previous studies.

#### 1.2. Aviation and Air Quality

To date, few studies have addressed the effects of aircraft emissions beyond airports on surface air quality, and nearly all have done so via the utilization of CTMs.

Both *Brasseur et al.* [1996] and *Köhler et al.* [2008] estimated, through the use of CTMs, that aviation increased surface ozone. *Köhler et al.* [2008] estimated that increases in surface ozone in the presence of aircraft may be due to vertical transport since the lower troposphere is a chemical sink of ozone. However, *Whitt et al.* [2011] isolated the impact of vertical transport alone from cruise altitude to the surface and found that a passive tracer took 62 to 77 days for its surface-to-cruise altitude fraction to exceed 0.5. They also found that the globally averaged vertical mixing time scales of cruise-altitude emissions were much longer than the wet removal time of 4 to 5 days for aerosol particles emitted in the lower troposphere. Thus, they show that vertical transport of cruise-altitude emissions to the surface, while affecting surface values, may be a lesser mechanism of increasing surface pollution outside the tropics than airport and landing/takeoff emissions.

Tarrasón et al. [2004], Barrett et al. [2010], Lee et al. [2013], Jacobson et al. [2013], and Morita et al. [2014] modeled the effects of cruise-altitude emissions on surface air quality, each providing a marginally different result, largely from differences in methodology and interpretation of results. Tarrasón et al. [2004] used a CTM to find that  $NO_X$  emissions above 1 km increased maximum surface ozone over Europe by 0.4 to 0.6 ppbv and ~1% in summer. They concluded that primary changes in surface particulate matter were negligible (<0.01%) relative to other emission sources and did not include them explicitly in model calculations. Barrett et al. [2010], on the other hand, used the Goddard Earth Observing System (GEOS)-Chem CTM to show cruise-level emissions accounted for ~80% (8000 deaths/yr) of the premature mortality impact of aviation, primarily as a result of secondary aerosol formation.

Lee et al. [2013] used the Community Atmosphere Model with Chemistry (CAM-chem) CTM to find that globally, aviation emissions increased regional surface ozone by up to  $\sim$ 1–2 ppbv in January and 0.5 ppbv in July, with the majority of perturbations coming from cruise emissions. Emissions above landing and takeoff altitudes ( $\sim$ 1 km) increased PM<sub>2.5</sub> by  $\sim$ 0.5% (<0.2  $\mu$ g/m³) over the United States, Europe, and eastern Asia. However, even where changes were statistically significant, the perturbations were less than 1% of background PM<sub>2.5</sub>. Mortality calculations were not reported due to uncertainties associated with the relatively small PM impact. Jacobson et al. [2013], using a CRM instead (GATOR-GCMOM), found global 2006 aircraft increased surface ozone by  $\sim$ 0.4% ( $\sim$ 0.05 ppbv) and surface PM<sub>2.5</sub> by  $\sim$ 83 ng/m³. All-altitude aircraft emissions were further estimated to increase human mortality worldwide by  $\sim$ 620 ( $\sim$ 240 to 4770) premature deaths/year, with half due to ozone and the rest to PM<sub>2.5</sub>. Similarly, Morita et al. [2014], using the NASA Goddard Institute for Space Studies (GISS) ModelE2 chemistry-climate model (here, run as a CTM), found that 2006 aviation emissions resulted in a 3 ng/m³ global increase in surface PM<sub>2.5</sub>, corresponding to 405 (182 to 648) premature mortalities per year.

While these studies have illuminated the relative contribution of aviation on surface air quality, each study differs in its focus on cruise- or all-altitude emissions, reported quantities of interest, or methodology for calculating mortality. This study uses many of the same models mentioned above but harmonizes model input where possible and provides further analysis of results.

# 2. Methods

Five models are used to run simulations at the following institutions (see Table 1): the GATOR-GCMOM model at Stanford University, GEOS-5 model at the NASA Goddard Space Flight Center (GSFC), the NASA GISS ModelE2 at Yale University, the CAM5 model at the University of Illinois at Urbana-Champaign and the National Center for Atmospheric Research (NCAR), and the GEOS-Chem model at the Massachusetts Institute of Technology (MIT). The models can be broadly classified either as climate-response models (CRMs; not to be confused with "cloud-resolving models") or as chemical-transport models (CTMs). As

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Institution	Model	Brief Description <sup>a</sup>	Simulation Years (Spin-Up) <sup>b</sup>
Stanford	GATOR-GCMOM	CRM: Fully coupled gas, aerosol, chemistry with model-calculated meteorological, cloud, and radiative feedbacks; uses subgrid chorded AC emissions	20 (0)
NASA GSFC	GEOS5-Replay (chem)	CTM <sup>C</sup> : AGCM coupled to stratosphere-troposphere chemistry module; replayed to MERRA reanalysis	1 (1)
	GEOS5-GOCART (aerosol)	CRM: AGCM coupled to GOCART aerosol module; AC effects feed back to model-calculated meteorology	1 (0)
Yale	NASA GISS ModelE2 fixed met. fields	CTM <sup>c,d</sup> : Coupled chem/aerosol with met. calculated by the model; AC effects do not feed back to met.	10 (2)
	Coupled met. fields	CRM: AC effects feed back to model-calculated meteorology	3 (19)
U. Illinois U-C, NCAR	CAM5 fixed met. fields	CTM mode: Coupled chem/aerosol; using MERRA met. fields; AC effects do not affect meteorology	1 (6)
	CAM5 Coupled	CRM mode: AC effects feed back to model-calculated meteorology	6 (6)
MIT	GEOS-Chem	CTM: Uses MERRA met. fields; coupled aerosol/chemistry	1 (5)

<sup>&</sup>lt;sup>a</sup>CRM = climate-response model; CTM = chemical-transport model; met. = meteorology; AC = aircraft.

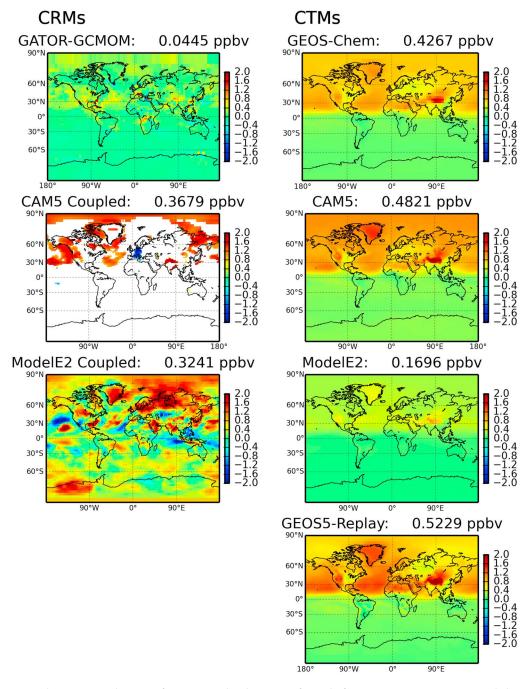
defined here, models that couple chemical and aerosol changes with online calculations of meteorological fields are called CRMs. CTMs either specify or strongly constrain dynamics with the use of meteorological data sets calculated off-line. The CRMs in this study, GATOR-GCMOM, CAM5 (CAM5 Coupled), ModelE2 (ModelE2 Coupled), and the GOCART configuration of GEOS-5 (GEOS5-GOCART), calculate their own meteorology, with aviation effects feeding back to the meteorological calculations. GEOS-Chem and a separate uncoupled CAM5 simulation, running as CTMs, are driven by NASA's Modern-Era Retrospective Analysis for Research and Applications (MERRA) reanalysis meteorological fields [Rienecker et al., 2011]. For surface ozone, GEOS5 runs in "Replay" mode (GEOS5-Replay), which incrementally adjusts the model dynamical fields toward the MERRA reanalysis [Rienecker et al., 2008]. ModelE2 is a chemistry-climate model capable of calculating meteorology but is run in this project both as a CRM and in a CTM mode, in which the same model-calculated meteorological fields are used to run simulations with and without aircraft. For this application, GEOS5-Replay and the uncoupled configuration of ModelE2 are also labeled as CTMs.

For aircraft emissions, models use the 2006 AEDT aviation emissions inventory of over 30 million commercial aircraft flights worldwide [Wilkerson et al., 2010]. GATOR-GCMOM treats emissions in a subgrid plume using chorded data from each of the individual flights, while other models used the same inventory, with emissions aggregated into 1° by 1° hourly grids. For other anthropogenic emissions, including CO, NO, NH<sub>3</sub>, SO<sub>2</sub>, OC, BC, and NMVOCs, models use the Intergovernmental Panel on Climate Change Fifth Assessment Report gridded emission inventory for 2005 assuming the Representative Concentration Pathways 4.5 emission trajectory [Clarke et al., 2007; Meinshausen et al., 2011]. Natural emissions, including biogenic, biomass burning, and lightning  $NO_X$  emissions, are either prescribed or calculated within each model based on validation from previous studies. GEOS5-Replay, GEOS5-GOCART, and both CAM5 and ModelE2 simulations are run at 2° latitude imes 2.5° longitude horizontal resolution; GEOS-Chem and GATOR-GCMOM simulations are run at 4° latitude  $\times$  5° longitude resolution. Vertical resolution varies for each model, with 25 to 43 layers in the bottom 15 km. Whereas the CRMs predict their own meteorology, the CTMs use MERRA reanalysis fields to drive meteorology. In this study, the uncoupled ModelE2 CTM uses the same model-calculated meteorological fields to run simulations with and without aircraft, though results using MERRA fields (not shown) were quite similar. Models are run for a number of simulation years (Table 1), recycling the same 2006 AEDT emissions and 2005 AR5 boundary conditions each year. The results, representing the changes in 2006 surface air guality from emissions, are compared. Table 1 briefly summarizes the models; more detailed descriptions of GATOR-GCMOM [Jacobson et al., 2011, 2013; Global Forecast System, 2013; Whitt et al., 2011; Jacobson, 2002, 2003, 2012; Jacobson and Streets, 2009; European Commission, 2014], GEOS-Chem [Murray et al., 2012,

Refers to the number of years over which results are averaged, following spin-up years (in parentheses, not included in the averages), until modeled results converge. Simulation years repeat 2006 AEDT aircraft emissions and AR5 anthropogenic emissions every year. MERRA 2006 meteorology (CTMs only) is used per model descriptions in the supporting information.

See section 2 for the CTM definition used here. GEOS5-replay allows for limited feedbacks.

dChemistry-climate model run in this project in both a CTM mode and a CRM mode.



**Figure 1.** Change in annual mean surface ozone (ppbv) due to aircraft. Results from GATOR-GCMOM, CAM5 Coupled CRM, and ModelE2 Coupled CRM are 20, 6, and 3 year averages, respectively; GEOS-Chem, CAM5 CTM, and GEOS5-Replay are 1 year averages after spin-up; ModelE2 CTM results are 10 year averages (see Table 1). Only statistically significant Coupled CAM5 values are displayed; all values are used to calculate global averages.

2013; Eastham et al., 2014; Fountoukis and Nenes, 2007; Guenther et al., 2006; Guenther and Wiedinmyer, 2007; Sakulyanontvittaya et al., 2008; Hudman et al., 2012; Jacob and Bakwin, 1991], CAM5 [Lamarque et al., 2012; Liu et al., 2012; Easter et al., 2004; Emmons et al., 2010; Guenther et al., 2006; Price et al., 1997], NASA GISS ModelE2 [Schmidt et al., 2014; Unger et al., 2010; Koch et al., 2006, 2009; Myhre et al., 2013; Shindell et al., 2013a, 2013b; Stevenson et al., 2013; Gery et al., 1989; Stockwell et al., 1997; Houweling et al., 1998; Bian et al., 2003; Koch and Hansen, 2005; Miller et al., 2006; Rayner et al., 2006; Price et al., 1997], and GEOS-5 [Rienecker et al., 2008;

<b>Table 2.</b> Change in O <sub>3</sub> and PM <sub>2.5</sub> , and Background AOD <sup>a</sup>					
Model	Average Ozone (ppbv)	Average PM <sub>2.5</sub> (μg/m <sup>3</sup> )	Background AOD <sup>b</sup>		
GATOR-GCMOM	0.045 (-2.5-4.9) 0.34%	0.0772 (-8.9-6.5) 0.42%	0.159		
GEOS-5	(Replay) 0.52 (0.28-1.6)1.92%	(GOCART) -0.17 (-1.8-2.3) -1.86%	0.091		
NASA GISS ModelE2	0.17 (0.13-0.22) 0.53%	0.0062 (0.004-0.008) 0.42%	0.161		
Fixed met. coupled	0.324 (-0.09-1.00) 1.15%	0.0165 (-0.015-0.071) 1.12%			
CAM5	0.48 (0.32-0.67) 1.80%	0.0034 (0.0014-0.007) 0.21%			
Fixed met.coupled	0.37 (0.12-0.83) 1.38%	0.0133 (-0.022-0.039) 1.18%			
GEOS-Chem	0.43 (0.27–0.65) 1.63%	0.0070 (0.0018–0.014) 0.14%	0.121		

 $<sup>^{</sup>a}$ O<sub>3</sub> and PM<sub>2.5</sub> changes are due to the effects of 2006 aviation emissions relative to an atmosphere with no aviation emissions. Values are 2006 annual global averages, with monthly averaged minimum and maximum values in parentheses. Relative change (%) in the global, annual value is shown for average ozone and PM<sub>2.5</sub>.  $^{b}$ Global average clear-sky aerosol optical depth (AOD) at 550 nm without aviation emissions. GEOS-Chem is total-sky

Duncan et al., 2007; Oman et al., 2011; Chin et al., 2002, 2014; Colarco et al., 2010; Guenther et al., 1995], and emissions are given in the supporting information.

While attempts at harmonization are made wherever possible, this project does not attempt to alter the models' best practices or configurations that have been validated in previous studies. For instance, while anthropogenic emissions are uniform across models, natural emissions (e.g., biogenic, biomass burning, and lightning  $NO_X$ ) may be prescribed, parameterized from dynamics, or calculated online during each simulation; synchronizing such emissions is not trivial and would likely introduce unforeseen variation that outweighs the benefits of intermodel regularization. While natural emissions, aerosol representations, model grid resolution, and simulation convergence criteria are not harmonized here, such treatment does remain constant within an individual team's simulations with and without aircraft emissions. We note that other model intercomparisons employ similar strategies [e.g., Lamarque et al., 2013; Søvde et al., 2014; Makar et al., 2015a, 2015b; Im et al., 2015a, 2015b; Brasseur et al., 2016]. In order to minimize but not eliminate intermodel differences, all  $PM_{2.5}$  and  $O_3$  results are displayed as the difference between simulations with and without aircraft emissions. This additionally highlights the small-in-magnitude changes in  $PM_{2.5}$  and  $O_3$  from aviation, shown here to be several orders of magnitude lower than ambient concentrations.

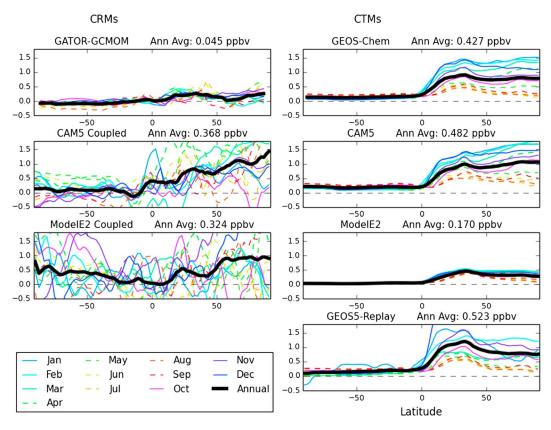
#### 3. Results

Results are analyzed as the difference between simulations with and without 2006 aircraft emissions. In plots and tables, perturbations are reported with spatially and temporally averaged perturbations. While this is the most straightforward metric for reducing model dimensionality and comparing changes across models, we note that averaging may artificially smooth perturbations and extreme events, diminishing peak values that may be more critical to air quality.

#### 3.1. Surface Ozone

Figure 1 compares the 2006 annual average change in surface-layer ozone due to aviation among the models. The left column shows results for GATOR-GCMOM, CAM5, and ModelE2 when run in CRM mode, averaged over 20, 6, and 3 years, respectively. In CRM mode, aircraft emissions feed back to radiation fields, temperature, atmospheric stability, clouds, winds, natural emissions, and transport processes between simulations with and without aviation. Note that while only statistically significant CAM5 Coupled values (see section 3.4) are displayed in Figure 1, all results are used to calculate the averaged values reported in tables, figures, and text. The global annual surface perturbations, given also in Table 2, are 0.34 to 1.4% (CRMs) and 0.5 to 1.9% (CTMs) of background ozone (~12 to 32 ppbv). The right side of Figure 1 shows results for models run as CTMs: GEOS-Chem, CAM5, ModelE2, and GEOS5-Replay. In these models, changes in ozone are not influenced by second-order changes in vertical mixing, removal rates, or radiation fields stemming from aircraft emissions; as a result, the perturbation fields appear "smoother" relative to the CRMs. Longer CRM simulations dampen this noise by averaging the interannual and seasonal variation, leaving a more distinct aviation response, discussed further in sections 3.2.3 and 3.4.

<sup>&</sup>lt;sup>o</sup>Global average clear-sky aerosol optical depth (AOD) at 550 nm without aviation emissions. GEOS-Chem is total-sky AOD. MODIS satellite clear-sky AOD (2001–2005) is 0.176. Dashed lines indicate background AOD values are not available.

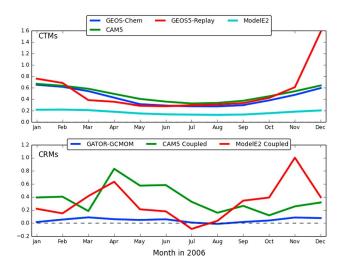


**Figure 2.** Change in monthly zonal mean surface ozone (ppbv) due to aircraft. Monthly values are averaged over the simulation length (Table 1).

The CTMs show that aviation emissions cause positive increases in the surface layer ozone predominantly in the Northern Hemisphere. The response is largest in high-altitude regions and in large-scale subsidence regions in the subtropics, with prominent maxima of 1.97, 2.42, 1.08, and 2.80 ppbv over the Tibetan Plateau for GEOS-Chem, CAM5, ModelE2, and GEOS5-Replay, respectively. There is a poleward extension of ozone over the North Atlantic and Greenland. Outside of the Tibetan Plateau, though differences everywhere are less than 1.5 ppbv. Aviation emissions are less abundant in the Southern Hemisphere, and the resulting surface response appropriately low (<0.5 ppbv) in all locations. In Europe and eastern North America, where population and aircraft emissions are particularly dense, the surface ozone perturbations are smaller. Over much of Europe (30° to 60°N and -15° to 35°E) annual perturbations are 0.5–0.9, 0.5–1.0, 0.3–0.6, and 0.6–1.3 ppbv in GEOS-Chem, CAM5, ModelE2, and GEOS5-Replay, respectively; over eastern North America (25° to 50°N and -95° to -65°E), perturbations are 0.5–1.0, 0.3–0.5, and 0.4–1.4 ppbv.

The annual ozone features of the CTMs suggest that in these models, horizontal transport and subsidence are the dominant processes bringing ozone and its precursors from above the landing and takeoff region to the surface. The resemblance between the difference field and background ozone (Figure S5) indicates that the increases in surface ozone occur chiefly beneath the primary subsidence pathways. Furthermore, despite the abundant air traffic and emissions released over Europe and eastern North America, the response in these regions is approximately half of the response over the subsidence regions, again demonstrating the large-scale spread of perturbations over a local response.

Like the CTMs, the CRMs show positive ozone perturbations off the North American coastlines and above the Tibetan Plateau, though local maxima and minima are also seen in several other locations. Coupled CAM5 maximum perturbations (+2.64 ppbv) are seen over the Tibetan Plateau and Eastern Russia, while minimum values (-3.17 ppbv) are almost exclusively over central Europe. There are no statistically significant positive ozone perturbations over central Europe in the CAM5 CRM results, and no significant results over eastern North America. Coupled ModelE2 maximum (+2.81 ppbv) and minimum (-2.27



**Figure 3.** Change in global monthly mean surface ozone (ppbv) due to aircraft.

ppbv) perturbations both appear west of North America over the Pacific Ocean with additional local maxima concentrated in the northern latitudes. GATOR-GCMOM shows maximum (+4.88 ppbv) and minimum (-2.53 ppbv) annual surface ozone perturbations over the Mediterranean Sea and southern tip of Greenland, respectively. The GATOR-GCMOM simulation results in smaller perturbations than other CRMs as noise is averaged across more years and also exhibits less downward transport of aircraft emissions to the surface. Additionally, the subgrid plume treatment of emissions in GATOR-GCMOM captures spatial variation in emissions

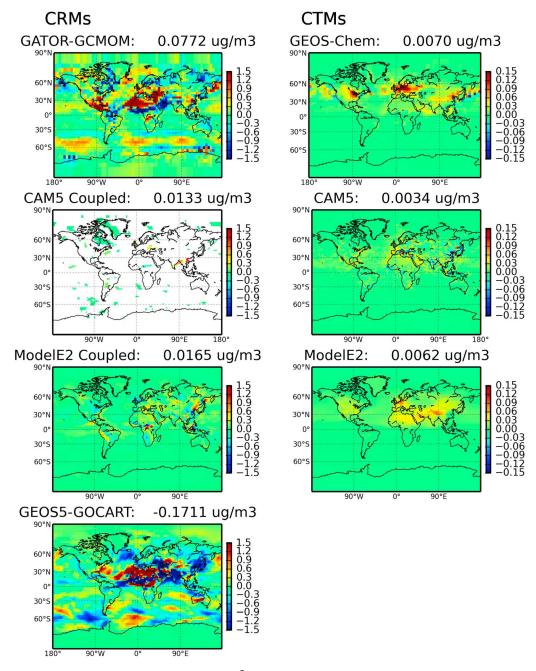
and associated chemistry that other models releasing emissions to the grid scale do not [Jacobson et al., 2013; Cameron et al., 2013].

Figure 2 shows the change in zonal surface ozone due to aviation, averaged over each simulation month. The CAM5 and ModelE2 coupled simulations show higher monthly variation with latitude, relative to their CTMs. Most models show little to no change in surface ozone in the Southern Hemisphere. Figure 3 shows globally averaged changes in surface ozone by month in 2006 for each model, with CTMs showing similar peaks during winter and lower values during the summer. These seasonal trends in perturbations can be affected by many atmospheric phenomena that are difficult to disentangle from one another. Helmig et al. [2007] found that ambient surface ozone measurements from four Arctic stations peak from December to May, which may contribute to the higher Arctic perturbations seen in the CTMs and, to a lesser extent, the CRMs. Whitt et al. [2011], on the other hand, show that vertical transport of aviation emissions from cruise altitude to the surface is enhanced in the Northern Hemisphere summer and dampened in winter. Their analysis indicates that surface ozone perturbations from aircraft emissions could be higher in summer, as more ambient stratospheric and aircraft-induced ozone is transported to the surface. Additionally, aircraft emissions can result in an increase of cruise-altitude air temperatures, increasing atmospheric stability and decreasing vertical transport [Jacobson et al., 2013]. As the CTMs do not change meteorological fields in simulations with and without aircraft, this effect would only appear in the CRMs. Despite the differences in seasonal aircraftinduced perturbations between the models, the global perturbations are generally less than the seasonal changes in ambient surface ozone; in winter months, ambient ozone is ~5 ppbv lower than in summer months, indicating that globally, the monthly surface ozone perturbations (less than 2 ppbv) fall within the seasonal variation of the CTMs' background fields. However, it is important to note that averages, while useful for intermodel comparison, smooth daily and regional variations that affect air quality.

# 3.2. PM<sub>2.5</sub>

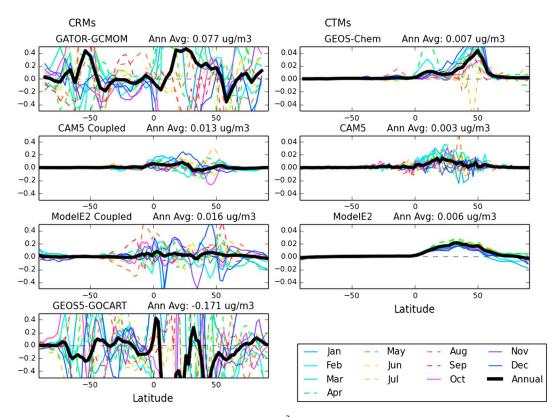
# 3.2.1. Feedback Effects on PM<sub>2.5</sub>

Figures 4 and 5 show the change in surface-layer  $PM_{2.5}$  due to 2006 aviation emissions for all models. Figure 4 gives annual average differences (with minus without aviation) at the surface. Figure 5 shows annually and monthly averaged differences by latitude. GATOR-GCMOM, CAM5 coupled CRM, ModelE2 coupled CRM, and GEOS5-GOCART allow aviation gases and aerosols to feed back to temperatures, temperature profiles, winds, clouds, and other meteorological variables, all of which affect the concentrations of nonaircraft particles and gases. One such mechanism is the influence of aviation aerosol on radiation, which can cause a small increase in near-surface wind speed, which in turn increases the emission rates of soil dust, sea spray, pollen, spores, bacteria, and biogenic gases, all of which contribute to  $PM_{2.5}$ . Similarly, changes in aerosols can change cloudiness via the aerosol indirect effect, causing a change in lightning- $NO_X$  emissions, precipitation, temperature, and air mixing that feeds back to ozone and particle formation and removal. *Wang et al.* [2015]



**Figure 4.** Change in annual mean surface  $PM_{2.5}$  (µg/m<sup>3</sup>) due to aircraft. Results from GATOR-GCMOM, CAM5 Coupled CRM, ModelE2 Coupled CRM, and GEOS5-GOCART are averaged over 20, 6, 3, and 1 year, respectively; GEOS-Chem and CAM5 CTMs are 1 year averages after spin-up; ModelE2 results are 10 year averages (all spin-up times listed in Table 1). Only statistically significant Coupled CAM5 values are displayed; all values are used to calculate global averages. Note the change in scale.

also compare model results running with and without feedbacks and show that aerosol indirect effects on clouds, temperatures, photolysis, and mixing are the primary process affecting modeled surface chemistry and PM<sub>2.5</sub>. A multimodel comparison of models running with and without feedbacks [*Makar et al.*, 2015b] makes the same conclusion. Furthermore, they show that the various representations of these feedbacks between models can produce a larger variation than the presence of feedbacks alone, emphasizing the need to further evaluate these interactions.



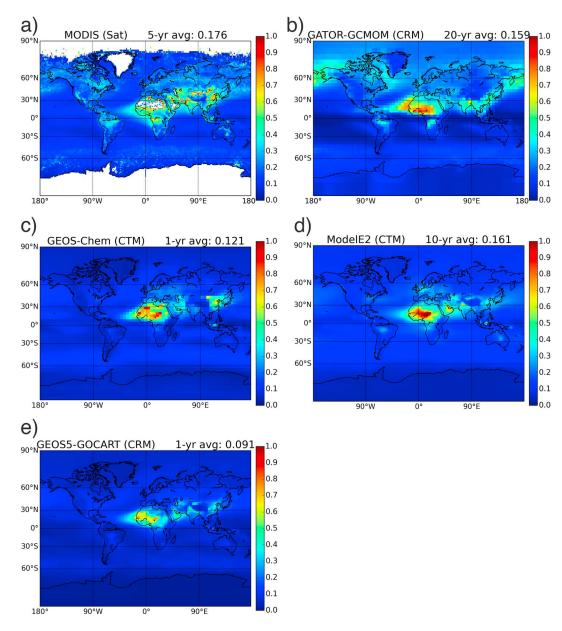
**Figure 5.** Change in monthly zonal mean surface  $PM_{2.5}$  ( $\mu g/m^3$ ) due to aircraft. Monthly values are averaged over the simulation length (Table 1). Note the change in scale between the CRMs and CTMs.

Sensitivity tests with GEOS5-GOCART wherein the interaction between radiation and the evolving aerosol field is removed, leaving only the background climatological aerosol field to interact with radiation, further support the importance of feedbacks. Here the aviation-induced perturbed aerosol field is decoupled from the circulation. The results (Figure S1) resemble those of the CTM responses, even when the radiation field feeds back to meteorology. This work helps further explain the different results of CTMs versus fully interactive CRMs. Additionally, it argues for the importance of including the feedback between radiation and the circulation in simulations involving aerosols, in particular those involving absorbing black carbon.

Omitting many of these interactions in the CTMs disentangles the primary aircraft emissions from the feedback effects on ambient PM, resulting in less monthly variation (Figure 5; note the change in scale between CRMs and CTMs). This is particularly evident in the ModelE2 and CAM5 coupled and uncoupled simulations. While the variation in the coupled CAM5 simulation causes many of the smaller perturbations to be statistically insignificant, the remaining values are generally greater in magnitude than those of the CTM at the same location and could indicate extreme events relevant to air quality calculations (Figure 4; note the change in scale between CRMs and CTMs).

# 3.2.2. Influence of Background Aerosol Fields

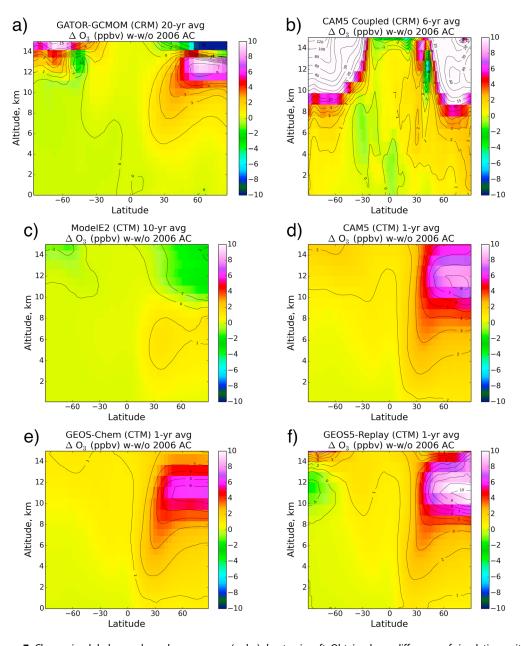
As a step toward determining the realism of the models' ambient aerosol fields, Figure 6 compares MODIS satellite-derived 550 nm aerosol optical depth (AOD) with predictions from GATOR-GCMOM, GEOS-Chem, ModelE2, and GEOS5-GOCART. AOD, a measure of light extinguished by aerosols in the column above the Earth's surface, can be used to estimate its relationship to surface PM<sub>2.5</sub> concentrations with varying complexity [Hoff and Christopher, 2009]. For instance, van Donkelaar et al. [2010] used AOD with a CTM to model surface PM<sub>2.5</sub> giving an uncertainty of 6.7  $\mu$ g/m³, when errors are weighted by population across the globe. Though a correlated modeled- and satellite-derived AOD field does not necessarily imply accurate surface PM<sub>2.5</sub> [Duncan et al., 2014], it does demonstrate a model's ability to capture total-column aerosol load, likely resulting in a more-accurate surface PM<sub>2.5</sub> distribution. Figure 6 shows that MODIS and the four models exhibit maximum aerosol loadings over the Saharan desert and Southeast Asia, consistent with areas of high dust



**Figure 6.** Mean ambient 550 nm aerosol optical depth (AOD). Results from (a) MODIS satellite clear-sky measurements (2001–2005 5 year average), (b) clear-sky GATOR-GCMOM CRM, (c) total-sky GEOS-Chem CTM, (d) clear-sky ModelE2 CTM, and (e) clear-sky GEOS5-GOCART CRM. All figures show background 550 nm AOD values without aircraft.

emission. Globally, the modeled AOD averages are within a factor of 2 and qualitatively resemble the MODIS retrievals, indicating that while the relative  $PM_{2.5}$  response to aviation varies greatly among the models, the background surface aerosol fields are generally well represented.

The difference in which background  $PM_{2.5}$  components are interactively modeled, however, varies significantly between the models, primarily from the inclusion of soil dust and sea spray (see Figure S6). The annual average  $PM_{2.5}$  surface concentrations in the GATOR-GCMOM CRM, the GEOS5-GOCART CRM, and the GEOS-Chem CTM, all of which include soil dust and sea spray, are 18.4, 9.2, and 5.1  $\mu$ g/m³, respectively, with peaks over the Saharan desert and Southeast Asia. Global surface perturbations from aircraft are 0.42%, -1.9%, and 0.14% of the background surface field (Table 2). Both CAM5 and ModelE2 simulations remove soil dust and sea spray components from perturbed aerosol fields and thus have a much lower background  $PM_{2.5}$  field from which aviation-induced changes can stem. Removing these components, perturbations of 1.13



**Figure 7.** Change in global annual zonal mean ozone (ppbv) due to aircraft. Obtained as a difference of simulations with and without 2006 aircraft (AC) emissions for (a) GATOR-GCMOM, 20 years, (b) CAM5 coupled CRM, 6 years, (c) ModelE2 CTM, 10 years, (d) CAM5 CTM, 1 year, (e) GEOS-Chem CTM, 1 year, and (f) GEOS5-Replay CTM, 1 year with spin-up times listed in Table 1. Model horizontal resolutions in Figures 7b–7d and 7f are  $2^{\circ} \times 2.5^{\circ}$ ; resolutions in Figures 7a and 7e are  $4^{\circ} \times 5^{\circ}$ .

(CAM5 coupled CRM), 1.60 (CAM5 CTM), and 1.47 (ModelE2 CRM and CTM)  $\mu$ g/m³ yield global perturbations from aircraft of 1.2%, 0.21%, 1.12%, and 0.42%, respectively. Thus, while the relative percentage change in PM<sub>2.5</sub> is similar with and without the inclusion of sea spray and soil dust, the overall magnitude change ( $\mu$ g/m³, as in Figure 4) is quite different. It should be noted that while all teams do represent dust and sea spray in their aerosol models, removing the components in postprocessing produces the significant differences discussed here. ModelE2, for instance, includes aviation-induced changes to sulfate and nitrate formed on top of natural dust in the perturbed PM<sub>2.5</sub> fields, though the magnitude is smaller than that of dust alone. Since CTMs do not include meteorological feedbacks, changes in background soil dust and sea spray are not modeled regardless of inclusion, and hence, perturbations remain smaller than those of the



CRMs. Conversely, GATOR-GCMOM and GEOS5-GOCART show large spatial changes in PM<sub>2.5</sub> where natural dust and sea spray emissions are abundant.

Results here can help explain the large variation in previous aviation mortality calculations. It is common to use PM<sub>2.5</sub> mass concentrations to calculate the health effects of PM<sub>2.5</sub>. Recent literature on the modeling and toxicity of individual particle components, however, indicates that individual PM<sub>2.5</sub> components play an important role as well [e.g., *Levy et al.*, 2012; *Li et al.*, 2016]. In omitting these two species, the CAM5 CRM and ModelE2 CRM results show the response of feedbacks on particulates not masked by abundant natural emissions, and modeling decisions related to these species could greatly change the number of aviation-induced mortalities. This demonstrates the importance in accurately representing interactions of aviation with the background aerosol field when calculating health effects. A study from *Im et al.* [2015b] similarly shows that dust and sea spray emissions heavily affect modeled surface PM<sub>10</sub> and PM<sub>2.5</sub> concentrations in CRMs and that further research is needed to identify the major uncertainties in PM model bias.

#### 3.2.3. PM<sub>2.5</sub> Response to Model Simulation Length in CRMs

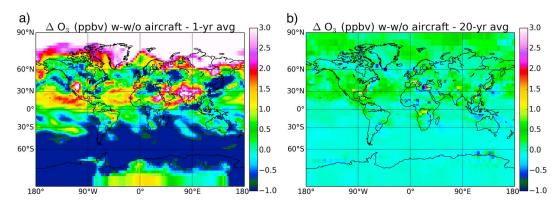
For the simulation lengths used in this study, predictions of surface-layer aerosols emitted by or produced in response to aviation are generally not consistent enough to diminish the variation that arises from coupling aerosols and dynamics. This is demonstrated in Figure 4, where the local CAM5 CRM surface results were not significant ( $\alpha$  = 0.05) over much of the globe. Over longer simulations, the aviation signal becomes more evident as noise diminishes with averaging. Among CRMs, CAM5, and ModelE2 Coupled simulations discard initial years as spin-up, removing some of the initial noise. GATOR-GCMOM does not discard initial model years (i.e., no spin-up) and instead obtains a 20 year average by repeating 2006 anthropogenic emissions and boundary conditions. GEOS5-GOCART also did not discard results, as the 1 year simulation is intended to demonstrate the magnitude of the aerosol-radiation-transport feedback in CRMs, and not necessarily to obtain a significant aviation response. It should also be noted that while not all surface values show perturbations above the 95% confidence level, the effects of aircraft at cruise altitudes are much more prominent, demonstrated in Figure 7 and consistent with previous work. It is anticipated that longer simulations could eventually yield more locations where confidence in surface perturbations surpasses 95% (see section 3.4), though it is unclear how long such simulations would need to be.

# 3.3. Cruise and Nonlanding/Takeoff Emissions

An important question arises as to the extent to which surface ozone and particles from aircraft are influenced by cruise-altitude emissions versus aircraft emissions in the boundary layer. To address this issue, additional simulations are performed with CAM5, GEOS-Chem, and ModelE2 CTMs that use only nonlanding/takeoff (non-LTO) emissions in the aviation emissions inventory, removing all emissions below 3000 ft (914 m). The resulting non-LTO surface  $O_3$  and  $PM_{2.5}$  perturbations are nearly identical to those of all-altitude emissions in Figures 1 and 4 but lower in magnitude (Figure S2). As with all-altitude emissions, the removal of LTO emissions results in positive global annual ozone and  $PM_{2.5}$  perturbations, with averages that are 90.1-100.4% and 42-88% of the baseline simulations, respectively. Thus, the LTO emissions contribute up to 10% of surface ozone and 12-38% of surface  $PM_{2.5}$  perturbations induced by all-altitude emissions in the CTMs. Additional perturbations are the result of emissions above LTO, which include emissions both above and below cruise altitude. However, even the maximum ozone and  $PM_{2.5}$  non-LTO perturbations are  $\sim 3.4\%$  (2.4 ppbv) and  $\sim 2.3\%$  (0.28  $\mu g/m^3$ ) of the 70 ppbv and 12  $\mu g/m^3$  U.S. Environmental Protection Agency (EPA) air quality standards.

In GATOR-GCMOM, the downward transport of cruise-level aerosols to the surface is minimal. Zonally and annually averaged number concentration differences at the surface (-16.5 to 22.8 cm<sup>-3</sup>) are within 4% of the peak cruise-level perturbation (573 cm<sup>-3</sup>). The large concentration difference alone indicates that the majority of particles created aloft are more likely to remain aloft than to descend to the surface layer and affect air quality (Figure S4). The slow vertical transport rate relative to the removal rate of particles is supported by *Whitt et al.* [2011]. GATOR-GCMOM additionally has a higher vertical resolution than the other models, particularly at cruise altitude where layers are on the order of 0.5 km versus 0.8–1.1 km, which may decrease the numerical diffusion of emissions to the surface.

Figure 7 compares zonal difference plots of ozone (ppbv) due to 2006 aviation from GATOR-GCMOM CRM, CAM5 coupled CRM, ModelE2 CTM, CAM5 CTM, GEOS-Chem CTM, and GEOS5-Replay CTM. The GATOR-



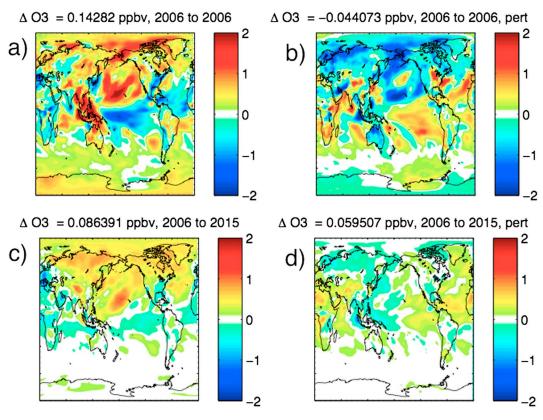
**Figure 8.** Change in GATOR-GCMOM global annual mean surface ozone (ppbv) due to aircraft for (a) 1 year at  $2^{\circ} \times 2.5^{\circ}$  horizontal resolution and (b) 20 years at  $4^{\circ} \times 5^{\circ}$ .

GCMOM results indicate a relatively small contribution of cruise-altitude ozone to the surface. The CAM5 coupled CRM shows larger differences in zonal ozone, with a local maximum over 40 ppbv at 71°N, 11.5 km altitude; the high ozone perturbations near the tropopause in CAM5 and GATOR-GCMOM CRMs are not produced by emissions but result from the displacement of stratospheric ozone due to aviation feedback effects on atmospheric stability and vertical mixing. The CTMs all show a transport of aircraftinduced ozone between cruise altitude and the surface, particularly near 30°N where zonal mean surface perturbations peak (see Figure 2, right column). Large-scale subsidence results in the downward transport of ozone and its precursors from aloft; however, it is clear that the majority of ozone perturbations occur aloft, not at the surface. Of the CTMs, ModelE2 in Figure 7c shows the smallest range of aviation effects on ozone, with a maximum perturbation of 1.28 ppbv at 87°N, 7.3 km, and minimum of -2.17 ppbv at 87°N, 14.4 km. The cause for the smaller response may be influenced by slightly coarser vertical resolution or different background ozone fields in ModelE2 but was not specifically identified here. The CAM5 model in CTM mode has maximum and minimum values of 8.27 and 0.17 ppbv at 82°N, 11.0 km, and 62°S at ground level, respectively. GEOS-Chem has a maximum perturbation of 6.54 ppbv at 84°N, 10.5 km, and minimum perturbation of 0.12 ppbv at 64°S at ground level. GEOS5-Replay has maximum and minimum values of 10.5 and -1.59 ppbv at 82°N, 11 km, and 88°S, 11 km, respectively. All models show little or no change in the Southern Hemisphere, and maximum ozone perturbations in most models are seen in the Northern Hemisphere, between 60-90°N and 9-13 km. ModelE2 shows a slight decrease in ozone from aircraft in that region, though differences are less than 2 ppbv.

While it is recognized that model grid resolution plays an important role in air quality simulations, global, multiyear simulations here necessitate the use of coarser grids than is ideal. Enhanced resolution allows for a more accurate representation of concentration gradients that are otherwise artificially smoothed over larger volumes [Arunachalam et al., 2011; Cameron et al., 2013; Jacobson, 2001; Li et al., 2016; Rissman et al., 2013; Schaap et al., 2015]. In this study, grid-scale volumes are several orders of magnitude larger than a young aircraft plume. Additionally, coarse vertical resolution may not accurately capture vertical mixing and may affect diffusion of emissions from cruise altitude to the surface. Resolving the need for both global and plume-scale coverage could reduce some of these uncertainties.

# 3.4. Temporal Averaging and Significance in CRMs

Figure 8 compares 1 year ( $2^{\circ} \times 2.5^{\circ}$ ) and 20 year ( $4^{\circ} \times 5^{\circ}$ ) averaged results for the differences in surface ozone (ppbv) from GATOR-GCMOM. The figures indicate that the maximum and minimum variations in ozone apparent during 1 year settle down significantly when averaged over 20 years. Part of the reason for this can be seen in Figure 9, which compares results from a random perturbation study from the CAM5-coupled CRM over 1 year versus 10 years. In the perturbed scenarios, the initial background temperature field is slightly altered by  $10^{-14}$  °K in order to measure the internal variability of the model using identical emissions and model treatment. This technique is used to generate an ensemble of simulations and provide criteria for significance testing. Differences between perturbed simulations give a defined threshold for the noise level in the model results. Where differences in ozone from the simulations with and without aircraft exceed the



**Figure 9.** Differences in CAM5 coupled CRM global annual mean surface ozone (ppbv). (a) Due to aircraft (AC) emissions for 1 year, (b) AC with perturbation-AC without perturbation for 1 year, (c) difference of simulations with and without aircraft for 10 years, and (d) AC with perturbation-AC without perturbation for 10 years.

noise from the random perturbation simulations of the same length, the differences in ozone are deemed statistically significant. Due to its relatively quick model run time, the CAM5 Coupled CRM is able to run ensemble simulations for significance testing, providing insight as to where results could be more reliable. Similarly, *Jacobson et al.* [2013] previously discussed the significance of aviation-induced changes in surface temperature using GATOR-GCMOM. Though not presented here, their results show that even small changes in surface temperature are statistically significant over ~26% and ~40% of the globe to a 95% and 85% confidence level, respectively.

Results in Figure 9 indicate that a random perturbation in the model causes deterministic chaotic variation that is larger in the first year than when averaged over 10 years. This chaotic variation is inherent in climate models due to variance that results from chaotic meteorology. A comparison of Figures 9c and 9d indicates that the globally averaged noise perturbation (0.06 ppbv O<sub>3</sub>) can be nearly the same (~2/3) as the signal (0.09 ppbv). As shown in Figures 1 and 4, the majority of small perturbations in the CAM5 coupled results are not significant at the 95% confidence level, though consistent peaks do remain. While each model will have its own significance criteria, this puts into perspective the results from the other CRMs. It is unlikely that Southern Hemisphere perturbations, for instance, are the result of a clear aircraft signal, rather than noise. Despite their inherent problem with chaotic variation, climate-response models are necessary for simulating the real feedbacks that occur between meteorology and chemicals in the environment. By decoupling these processes, CTMs are capable of producing results with much less computation and can be particularly valuable when time and resources are limited; however, implications from the results are also limited as they do not capture real interactions between emissions and circulation.

#### 3.5. Health Considerations

The global annual average aviation-attributable positive surface ozone and  $PM_{2.5}$  perturbations are ~0.7% (~0.5 ppbv) and ~1.5% (<0.2  $\mu g/m^3$ ) of the current 70 ppbv and 12  $\mu g/m^3$  U.S. EPA primary air quality



standards. However, these standards are both higher than the World Health Organization (WHO) recommendation of 100  $\mu$ g/m<sup>3</sup> (approximately 50 ppbv at STP) 8 h mean ozone and 10  $\mu$ g/m<sup>3</sup> annual mean PM<sub>2.5</sub> [World Health Organization, 2006], and investigations of mortality and morbidity attributable to both short-term and long-term ozone exposure have found no clear evidence of a no-effect threshold for ozone exposure [Ostro et al., 2006; Jerrett et al., 2009; Ji et al., 2011]. Similarly, when defining the integrated exposure response model for PM<sub>2.5</sub> used in an assessment of the WHO global burden of disease, the health consequences of ambient PM<sub>2.5</sub> are likely nonlinear and historically underestimated [Lim et al., 2013]. Given the flexible nature of regulatory standards and the lack of a lower exposure limit for impacts, direct comparisons of changes in exposure to a specific set of regulatory standards without reference to background conditions or population composition are therefore an incomplete estimator of risk.

### 4. Discussion

Five global models are intercompared to examine the impacts of 2006 commercial aircraft emissions worldwide on surface ozone and PM<sub>2.5</sub>. The models are run using different baseline and sensitivity cases, offering a comparison between climate-response models (CRMs), chemical-transport models (CTMs), and models that integrated aspects of both CRMs and CTMs. Where possible, meteorology, emissions, boundary conditions, and grid resolution are harmonized to minimize discrepancies in model results.

For the surface ozone exercise, the focus on harmonization results in very similar spatial distributions of aviation-induced surface ozone perturbations in the CTMs. This demonstrates the controlling influence of horizontal transport over chemistry in determining the changes in surface ozone due to aviation. This response is evident as well in the CRMs, despite the fact that the CRMs allow feedback between the transport circulation and the perturbations from the aviation emissions. In this sense, the CRMs support the basic character of the ozone response to aviation emissions seen with the much-less-resource-intensive CTM simulations. Globally, all models report an increase in annual surface ozone from aviation (0.34 to 1.4% in CRMs and 0.5 to 1.9% in CTMs) primarily in the Northern Hemisphere where most aircraft emissions are released.

The results from the aerosol analysis, which are more clearly different between CRMs and CTMs, have illuminated the modeling landscape in a very different but equally valuable fashion. The 1 year free-running GEOS5-GOCART experiments provide a link between the two classes of models: when radiative heating is limited to climatological aerosols, GOCART produces results very similar to the CTMs, but when the full aerosol field is taken into account, the GOCART results look much more like the GATOR-GCMOM results. The changes in surface-level PM<sub>2.5</sub> in the CTMs (0.14 to 0.4%) and CRMs (-1.9 to 1.2%) depend on varying background aerosol fields, feedbacks to nonaviation aerosols, and simulation length.

CAM5 Coupled results demonstrate that the modeled small aviation-induced perturbations, particularly in the Southern Hemisphere, are not likely to be statistically significant due to the chaotic variation inherent in CRMs. However, the noise-to-signal ratio in CRMs reduces over longer simulations, increasing the confidence in results. The CTMs, on the other hand, do not display the high spatial variability seen in the CRMs but exclude important feedbacks between aerosols, chemistry, and circulation that are relevant to assessing the impacts of aviation on air quality.

#### 4.1. Limitations and Future Research Needs

Presently, CTMs are more commonly used in modeling air pollution for regulatory purposes. Such models can include important reactions while remaining computationally less expensive than CRMs but do not include feedbacks to meteorology that are shown here to induce relatively large differences in results. However, "feedbacks" are difficult to disentangle from chaotic variations in meteorology, making attribution a challenge, and multiple approaches necessary. Recent results from the AQMEII-2 project have helped elucidate the effects and uncertainties associated with coupling air quality and meteorology. However, as the trend for models to include feedbacks increases, there remains a need for future research to investigate the response of these meteorological feedbacks and assess their computational cost and accuracy with regulatory utility. Models that incorporate feedbacks in an inexpensive way (like GEOS5 in Replay mode), or models that can be run in both CRM and CTM modes, (i.e., with and without feedbacks, such as CAM5 and ModelE2), may be particularly useful in bridging the gap between CRMs and CTMs specifically on shorter time scales and warrant further research.



Additionally, this study does not delve into the ability of each model to accurately assess the background atmosphere species relevant to air quality, including  $NO_X$ ,  $HO_X$ , ozone, ammonia, and particulate matter. Such a study would require an analysis of these species and their transport throughout the troposphere over time. Similarly, this study does not perform a comprehensive comparison of the effects of modeling decisions that could not feasibly be harmonized, including the composition and treatment of aerosols; distribution of natural emissions; and model spatial and temporal resolution. While beyond the scope of this intercomparison, a more in-depth analysis of these would be valuable in determining the nonfeedback causes for model differences. Currently, the small changes in air pollutants modeled here make it difficult to differentiate between the models. Comparisons with ambient conditions would help quantify the range of model errors and contextualize the results in terms of accuracy and statistical significance.

Future work that weighs the utility of meteorological feedbacks within a model against computational cost could be particularly valuable to the regulatory community. While the global grid resolutions used in this study are too coarse to accurately assess the regional effects of aviation on air quality, results from this study give a possible range of global aircraft-induced ozone and PM<sub>2.5</sub> perturbations. These ranges, wider from CRMs due to feedbacks and more degrees of freedom, contextualize previous studies' results and deepen our understanding of the health impacts of aircraft emissions.

#### Acknowledgments

This work was funded by the Federal Aviation Administration Office of Environment and Energy. The authors particularly thank Rangasayi Halthore for his continued support, and three anonymous reviewers for their valuable feedback. Any opinions, findings, conclusions, or recommendations expressed in this material are solely those of the authors and do not necessarily reflect the views of the U.S. FAA. Annually averaged surface-level pollutant fields for each team are available in the supporting information. Additional data used to create figures and tables are available from the corresponding author upon request.

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