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Coupled Chemistry-Climate Effects from 2050 Projected Aviation Emissions

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Abstract. Analyses of the climate effects of 2050 aviation emissions have been conducted with two coupled Chemistry Climate Models (CCMs) including experiments with coupled ocean models. The baseline 2050 aviation emissions scenario projects emissions ~5 times those in 2006. Simulations suggest a corresponding growth in the climate impact of aviation by 2050. Positive radiative forcing from contrails reaches +80mWm^{-2}. Enhanced upper tropospheric and lower stratospheric ozone (O_3) due to aviation nitrogen oxide (NOx) emissions causes a radiative forcing of +60mWm^{-2}. Changes in methane (CH_4) lifetime induced by aviation are estimated to cause -25mWm^{-2} of radiative forcing in 2050. Simulations indicate that moderate changes in water vapor emissions from changes in combustion efficiency will not have significant forcing. Non-linear effects due to particles (black carbon and sulfur) included in these calculations suggest an important role for black carbon (BC) in increasing contrail cirrus ice crystal number, leading to net warming. Sulfur emissions brighten clouds and provide a net cooling, but this is dependent on uncertain background sulfur levels. Thus alternative aviation fuels with reduced sulfur and BC may alter the future climate impact of aviation, but the sign is dependent on specific processes represented and the background state. Regional perturbations due to contrail and particulate emissions may result in statistically significant regional surface temperature changes in coupled model simulations in areas near or adjacent to flight corridors, but significant signals only emerge after 20-50 years of simulation. Many regions with high regional aviation forcing do not experience net surface temperature changes because of advective rather than radiative driving of temperatures. Surface temperature signals are not significant globally even in long coupled simulations. Short-lived non-uniform aviation forcing will thus affect climate differently than uniform forcing in the coupled climate system.

1. Introduction

Aviation has several important impacts on climate (Wuebbles et al., 2007; and Lee et al, 2009, Brasseur et al., 2008 & 2015). Commercial aircraft currently emit ~2% (Wuebbles et al., 2007) of global fossil-fuel related emissions of carbon dioxide (CO_2). Aircraft also emit water vapor (H_2O), nitrogen oxides (NO_x), various organic gases, sulfur dioxide (SO_2), black

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carbon (BC) soot, other organic matter, and particulate sulfate. These chemicals may alter upper level cloudiness, atmospheric composition, and climate in several different ways.

For low enough temperatures and high enough exhaust humidity, aircraft emissions create condensation trails, or contrails (Schmidt, 1941; Appleman, 1953). If the ambient air is supersaturated with respect to ice, linear contrails may persist for minutes to several hours (Minnis et al., 1998). The optical thickness of existing cirrus may be enhanced by the additional nuclei resulting from contrails that have aged due to spreading, shearing, and sublimating (Minnis et al., 1998). Aircraft emission effects on ozone (O_3) and methane (CH_4) lifetimes may also have radiative feedbacks on climate (Brasseur et al 2015).

Recent work summarized by Brasseur et al (2015) has shown significant 'non-linear' effects from aviation aerosols due to (a) aerosol absorption, termed 'direct' radiative effects and (b) modification of contrail and natural cloud microphysical properties by aerosols, or 'indirect' radiative effects. These effects may be as large or larger than the radiative effects of contrails themselves (Righi et al., 2013, Gettelman and Chen, 2013, Zhou and Penner, 2014). Aviation soot aerosol may change their own properties as they age (Heymsfield et al 2010). Soot inclusions within and between contrail and background cloud particles also create a cloud absorption effect by enhancing contrail burnoff (Jacobson et al., 2013).

Many studies have explored the total non-CO_2 (short lived) effects of aviation radaitive forcing. Lee et al. (2009) performed a review and meta-analysis and estimated aviation radiative forcing (RF) of 59 to 110 mWm^{-2} for Ozone, -52 to -28mWm^{-2} for CH_4, 37 to 55mWm^{-2} for linear contrails, -12 to -18 for SO_4 (direct) and 9 to 13mWm^{-2} for BC (direct). Chen and Gettelman (2016) recently performed simulations with one of the models used here and found significant indirect effects of aviation aerosols, especially sulfate, and RF increases by a factor of 5-7 from 2006 to 2050.

This manuscript evaluates and assesses the radiative forcing, ozone perturbation and surface temperature changes from projected levels of aviation emissions for 2050 in two advanced General Circulation Models (GCMs) with common scenarios for 2050 aviation emissions. The study focuses on simulations with coupled ocean, land and ice models. The goal is to determine statistically significant signals for small perturbations in coupled simulations. Previous studies have not been conducted with fully coupled "climate-response" models with small aviation forcing.

Section 2 describes the models, methodology and scenarios. Section 3 describes the results, and section 4 discusses them with a focus on comparing models. Section 5 provides conclusions and suggestions for future work.

2. Methodology

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The climate-related effects described here include the radiative effects of contrails, the chemical effects of NOx on atmospheric concentrations of ozone (O_3) and methane (CH_4), and the direct and indirect (or non-linear) effects of atmospheric particles resulting from emissions of BC, organic compounds, and sulfur species.

We use two different models with interactive chemistry and climate, described below.

2.1 GATOR-GCMOM

GATOR-GCMOM (Gas, Aerosol, Transport, Radiation, General-Circulation, Mesoscale, and Ocean Model) is a one-way nested global-through-urban atmosphere-ocean-land model [Jacobson et al., 2011, 2013]. Resolution is 4^o (latitude) x 5^o (longitude) horizontal resolution with 68 vertical sigma-pressure layers from 0-60 km (0.219 hPa), including 15 layers from 0-1 km and 500-m resolution from 1-21 km. The model is initialized January 1, 2050 with 1^ox1^o reanalysis meteorological fields and ocean surface temperatures from January 1, 2012 [*GFS*, 2013] and run forward with no data assimilation. A single temperature profile was used to uniformly scale the ocean temperature relative to the surface, with temperatures at depth a constant ratio to the surface temperature. Thus the sub-surface ocean is not in balance as the stratification does not reflect spatial variations.

GATOR-GCMOM explicitly treats both the microphysical and radiative effects of aerosols on clouds and precipitation (Jacobson, 2012). Clouds, radiation and chemistry are coupled together, with feedbacks between them. GATOR-GCMOM treats the sub-grid evolution of aircraft particle and gas exhaust, including sub-grid contrail formation from each individual flight (30 million annually) [Jacobson et al., 2011, 2013; Whitt et al., 2010]. Non-aircraft anthropogenic emissions of NOx, SOx, NH3, CO, NMVOCs, CH4, BC, OC for 2050 are derived from the Fifth Assessment Report (AR5) inventory for 2050 assuming the Representative Concentration Pathway (RCP) 4.5 trajectory [Clarke et al., 2007]. In addition, for the long-lived greenhouse gases, CO_2, N_2O, CFCs, and HFCs, the EDGAR 2010 inventory [European Commission, 2014] extrapolated to 2050 are used. Biomass-burning gas and particle emissions are as in *Jacobson* [2012]. Natural emissions from lightning, soils, oceans, and vegetation are calculated as a function of meteorology as in Jacobson and Streets [2009].

The following global simulations are run for GATOR-GCMOM:

1) 2050 climate, 2050 non-aircraft anthropogenic emissions, and 2050 chorded aircraft emissions.
2) Same as (1) but with 2006 chorded aircraft emissions (but 2050 climate and 2050 non-aircraft anthropogenic emissions).
3) Same as (1) but with no aircraft emissions.
4) Same as (1) but with no BC in aircraft emissions.
5) Same as (1) but with no SOx, NOx, CO, HC aircraft emissions.

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6) Same as (1) but no contrail optical properties (contrail microphysics still occurred)
7) Same as (1) but with zero sulfur and 50% black carbon emissions (low-sulfur fuel scenario).

The simulations above have been run six complete years we show the first 5 years for comparisons. Results are substantially the same if year 6 is included. 20-year results for 2006 chorded emissions (with 2006 anthropogenic emissions and 2006 meteorology) have been documented by Jacobson et al. [2013].

Due to the high internal variability of the coupled climate system, long simulations are needed to achieve equilibrium. The GATOR-GCMOM simulations are transient simulations and are not in equilibrium. Long timescale feedbacks will not have fully adjusting to the small aviation perturbation. This will limit interpretation of small surface temperature signals, but should not affect atmospheric responses to aviation ozone and aerosols. We recognize there are limits to this method. We use longer CESM simulations (see below) to understand the impact of these assumptions.

2.2 CESM

Community Earth System Model (CESM, Hurrell et al 2012) with version 5 of the Community Atmosphere Model (CAM5) (Neale et al., 2010) uses a full gas-phase chemical mechanisms including tropospheric and stratospheric chemistry with 133 species and 330 photochemical reactions (Lamarque et al., 2012). CESM includes a detailed treatment of cloud liquid and ice microphysics (Morrison and Gettelman, 2008), including particle size distributions, a detailed mixed phase with a representation of water uptake onto ice (the Bergeron–Findeisen process) and ice supersaturation (Gettelman et al., 2010). Heterogenous freezing on dust and homogenous freezing are represented. Cloud microphysics is coupled to a consistent radiative treatment of ice clouds, and a 7-mode Modal Aerosol Model (MAM7) (Liu et al., 2012) with particle effects on liquid and ice clouds (Liu et al., 2012).

Aviation emissions for water and other chemical species are put into CAM following Chen et al (2012) and using the AEDT emissions inventories (as described below). The contrail parameterization of Chen et al., (2012) is able to produce a reasonable spatial and seasonal distribution of contrails compared to observations (Chen et al., 2012). CESM has been used to estimate the present day impact of aviation emissions by Chen and Gettelman (2013). Gettelman and Chen (2013) estimated radiative effects of aviation aerosols. Chen and Gettelman (2016) estimated aviation RF in specified dynamics simulations without chemistry.

2.2.1 CESM Specified Dynamics Simulations

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To determine small radiative forcing perturbations, CAM specified dynamics (CAM-SD) simulations are used to eliminate meteorological noise. CAM-SD simulations use fixed meteorology (imposed pressure, winds and atmospheric and sea surface temperatures). Water, clouds, aerosols and chemical species are freely advected. The use of the SD model does not allow the feedback of aviation impacts or chemistry on the background meteorology.

Two sets of SD simulations were performed. The first set of SD simulations for determining contrail and aerosol effects are 10 years long, and use meteorology specified from a CESM coupled run. For future years, 4 sets of driving meteorology for each year (representing evolving forcing) were derived by running CAM with SSTs from a coupled simulation with RCP4.5 or RCP8.5 forcing for that year. No aviation emissions are included in the meteorology. Ensembles are created with a unit temperature perturbation. Then 4 pairs of simulations with and without aviation emissions are performed for each specified year using that meteorological forcing. Each of the 4 simulation pairs uses different meteorology, to test the variability of the results with respect to meteorology. The different years tested between 2006 and 2050 are: 2006, 2016, 2026, 2036 and 2050. For each of the 4 ensembles and each year (2006,2016,2026,2036,2050), we perform SD simulation pairs using RCP4.5 with different scenarios as outlined below.

SD simulations examining NOx effects are 5 year simulations are performed for the year 2050. These are basically the same as the 2050 CESM SD simulations, but run with more comprehensive chemistry. RCP4.5 boundary conditions of non-aviation NOx and volatile organic compounds (VOCs) were obtained from the IPCC RCP4.5 scenario for year 2050 (Van Vuuren et al., 2011). The monthly surface concentrations of longer-lived species, e.g., CO_2, CH_4, chlorofluorocarbons (CFCs), and nitrous oxide (N_2O), were specified as boundary conditions based on the IPCC RCP4.5 scenario. Meteorological fields to drive the SD simulation are also taken from a coupled CESM simulation for 2050.

The University of Illinois Urbana Champaign (UIUC) Radiaitve Transfer Model (RTM) was used offline to calculate the forcing associated with aviation NOx-induced short-term O_3, in CESM while contrail effects are calculated in-line. The UIUC RTM calculates the flux of solar and terrestrial radiation across the tropopause (e.g., Jain et al., 2000; Naik et al., 2000; Youn et al., 2009; Patten et al., 2011).

2.2.2 CESM Coupled Simulations

Coupled Simulations were also performed with CESM to explore potential surface temperature responses to aviation forcing. Coupled simulations feature a free running atmosphere coupled to a full dynamic ocean and sea ice model with a land surface model. Feedbacks between chemistry, clouds and meteorology occur. Simulations were performed for 50 years,

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starting from a spun up control run of CESM for 2050 with RCP4.5. We only use the last 20 years for analysis, and allow 30 years for the model to adjust. These runs are closer to equilibrium than the GATOR-GCMOM simulations.

In order to evaluate the effect of transients in the simulations we also perform shorter CESM coupled experiments. These transient CESM simulations use the same ocean initial condition as GATOR-GCMOM: GFS reanalysis for SST for Jan 1, 2012. A single temperature profile was used to uniformly scale the ocean temperature relative to the surface, following GATOR-GCMOM. Salinity was not changed from the standard initial conditions in the RCP runs (the GATOR-GCMOM salinity was unstable in CESM). The biggest difference is the uniform temperature profile scaling used in the ocean. This run is analyzed to show the effect of a non-equilibrium state on the results from GATOR-GCMOM.

2.3 AEDT Emissions/Scenario

Simulations are conducted using a common set of emissions scenarios. Emissions used in this study come from the Aviation Environmental Design Tool (AEDT) emission inventory (Barrett et al., 2010; Olsen et al., 2012; Brasseur et al., 2015). The AEDT dataset is an hourly inventory of global aircraft emission mass of ten emission species over a 1° x 1° latitude-longitude mesh with a vertical spacing of 150 m in the year of 2006. 2006 data are chorded (individual flight tracks), and 2050 data are scaled and gridded. For GATOR-GCMOM, individual flight track 2050 aircraft emissions are obtained by extrapolating 2006 emissions in the same flight paths to 2050 by scaling the current year 2006 flight data by the ratio of 2050 to 2006 emissions. Emission indices from Barrett et al., (2010) are used for different species to determine particulate emissions. Both models use greenhouse gas concentrations in 2050 from the Representative Concentration Pathway 4.5 scenario (RCP4.5).

Different scenarios are listed in Table 1. The Baseline scenario assumes no operational or technology improvements. Scenario 1 has reduced fuel burn because of assumed technology improvements. Scenario 2 assumes the same fuel burn of Scenario 1 with an 'Alternative Fuel' (Alt Fuel) that has no sulfur and 50% less BC (soot) emissions. Scenario 3 assumes Scenario 2 (reduced fuel burn with no sulfur and 50% BC) with +5% higher aviation H_2O emissions from the engine exhaust. These scenarios were implemented in CESM. GATOR-GCMOM used 2050 baseline emissions, and a modified scenario 2 that assumes fuel burn of the Baseline scenario and an Alt Fuel version that has no sulfur and 50% less BC. This scenario (run only by GATOR-GCMOM) is called 'Scenario 2B'.

The baseline emissions scenario has a fuel burn in 2050 that is 5 times that of 2006 emissions. Over E. Asia, the 2050 emissions are nearly 8 times those of 2006. 2050 emissions changes are lower over the U.S. (2 times 2006) and Europe (4 times 2006). The reduced fuel burn scenario (Scenarios 1 and 2) is only 3 times 2006 emissions, with 4 times 2006 Asia, and 2 times 2006 for the U.S. and Europe.

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The 2050 AEDT gridded inventory (Barrett et al., 2010; Brasseur et al., 2015) for Scenario 1 assumes a combined aircraft technology and operational improvement consistent with maintaining a 2 %/yr improvement in aviation system efficiency and a NO_x-related technology improvement consistent with published ICAO/CAEP scenarios to 2036 extended to 2050 based on NASA N+3 and N+4 targets of "better than 75%" (Barrett et al 2010). The Baseline scenario does not include the operational or technology improvements that were included in Scenario 1.

2.4 Simulations

GATOR-GCMOM was run for five model years with and without aviation emissions for baseline and Scenario 2B in a fully coupled configuration. Sensitivity tests were conducted as described in Section 2.1. CESM was run in both a specified dynamics (SD) mode, without feedbacks between emissions, chemistry and meteorology, to determine radaitive forcing (Section 2.2.1) and in a fully coupled mode to compare with GATOR-GCMOM (Section 2.2.2). CESM coupled experiments were run for 50 model years for longer-averaged statistics. CESM was run with baseline emissions for 2050 and all three scenarios.

2.5 Statistical Significance

Statistical significance was assessed using a 2-sided Student's t-test (e.g. Chervain and Schneider, 1976), estimating the standard deviation of annual means from the simulations with one degree of freedom for each year. Statistically significant differences are defined as those above 90% confidence. We also performed a paired sample t-test with autocorrelation on the difference of the annual means and got essentially the same significance results (with a more formal treatment of autocorrelation). The short nature of the GATOR-GCMOM simulations (5 years) means that the standard deviations may be large (and the degrees of freedom small). CESM simulations can be evaluated in a similar 5-year window, or with a longer 20-year window from the first 20 or last 20 years (31-50) of 50-year simulations to explicitly address the issue of short and transient simulations.

3. Results

First we present general results for fuel burn from the scenarios and illustrate the potential for contrail formation. We then examine the model-derived radiative forcing from contrails and other aviation emissions, including the radiative effects of chemical perturbations and the "non-linear" effects resulting from aircraft emissions of soot and sulfur. Next we examine the model-derived chemical effects on ozone and methane from NOx emissions. Finally, we analyze the resulting temperature response from the aviation perturbations in 2050 based on coupled simulations. Note that the effect of alternative fuel is Scenario 2B v. Baseline for GATOR-GCMOM and Scenario 2 v. Scenario 1 for CESM.

3.1 Radiative Forcing in CESM

Radiative forcing for contrails and from aviation-related aerosols (both direct and indirect effects) were assessed in CESM. Top of atmosphere net downward solar plus thermal-infrared irradiance changes (a surrogate for radiative forcing but not the same quantity) were assessed in both CESM and GATOR-GCMOM. For clarity we focus on CESM radiative forcing.

Figure 1 (adapted from Chen and Gettelman 2016) shows the evolution of the radiative forcing over time in CESM for different scenarios using SD simulations. The effects of water vapor only emissions (leading to contrail formation) are shown in Figure 1A, the effects of H_2O + BC + SO_4 emissions in Figure 1B and the effects of BC+SO_4 emissions only in Figure 1C, as the residual of panel B minus panel A. Note that since H_2O emissions for Scenario 1 and Scenario 2 are the same, their RF in Figure 1A is the same. Figure 1A shows clear increases in contrail cirrus radiative forcing to 2050. While global emissions grow by a factor of 5, contrail cirrus RF may grow by a factor of 7 (12 to 90 mWm^{-2}). Note however that a warmer planet, using RCP4.5 meteorology (solid lines in Figure 1), lowers the resulting aviation RF relative to what it would be using 2006 meteorology (dashed lines in Figure 1), due to a reduction in contrail formation conditions at warmer temperatures (Chen and Gettelman, 2016). This assumes aircraft fly at the same pressure altitude in 2050 as today.

RF grows faster than the emissions because the regional pattern of emissions is changing. Emissions are rising faster in regions like Asia, and in these regions the RF may be more sensitive to perturbations. Sensitivity may be higher either because there are fewer contrails in 2006 or the chance of forming contrails at flight altitudes is higher (since aircraft are always in the troposphere at low latitudes).

Figure 1C shows the impact of aerosols in CESM. In the scenarios with significant sulfur emissions (Baseline, red and Scenario 1, green) there is significant decrease in TOA radiative forcing from aerosols (Figure 1C) that is anticipated to cause a near-surface cooling. Each line represents an average of 4 simulations (spread shown as error bars with symbols at the ends in Figure 1A and 1B).

Eliminating sulfur in Scenario 2 (blue) and Scenario 3 (purple) eliminates the negative radiative forcing (Figure 1C). The net forcing is then positive (Figure 1B) and similar to figure 1A. Scenario 3 has a small amount of additional water vapor from aircraft. There is a small increase in contrail RF in 2050, on the order of 0-5% for a 5% increase in water vapor in Scenario 3, also seen in Figure 1B. The effect is small since most of the water in the contrails comes from the ambient atmosphere, not the fuel. The difference between scenario 2 and scenario 3 is not statistically significant.

Additional radiative forcing components are listed in Table 2, reprinted from Brasseur et al (2015). In particular, we have added CAM5 values of radiative forcing for 2050 for short-term effects of ozone, methane and water vapor for Scenario 2 (alternative fuels). We have also added available RF values for contrails and aerosols (Figure 1) from CESM-CAM5. Other values are as described in Brasseur et al (2015). New values are in bold, and show small effects of alternative fuels on minor species radiative forcing. The reduction in sulfur and black carbon for alternative fuels in Scenario 2 changes short-term ozone RF by +2.2mWm^{-2}, long-term ozone RF by -0.7 mWm^{-2}, methane by -2.2 mWm^{-2}, and water vapor by -0.5mWm^{-2}. These effects in CESM are minor.

3.2 2050 Aviation NOx-induced effects on Ozone

3.2.1. Effects on ozone

Figure 2A and B illustrate the simulated changes in zonally-averaged O_3 due to aviation emissions for (A) GATOR-GCMOM and (B) CESM. Note that CESM uses years 30-50 of a long coupled simulation while GATOR-GCMOM uses the first 5 years of a 5-year simulation. Results for CESM at different averaging periods are in the supplement, and are more similar between models.

Ozone radiative forcing is especially sensitive to the predicted ozone distribution in the Upper Troposphere and Lower Stratosphere (UTLS) (Lacis et al., 1990). GATOR-GCMOM (Figure 2A) projects a peak increase in ozone at cruise altitude between 45˚N and 90˚N of ~60 ppb which is not statistically significant. Effects at higher altitudes likely depend on perturbations to the temperature gradient altering transport. However, due to variability of the stratospheric circulation they are not significant. Significance is illustrated by the red dots in Figure 2 and other figures. Effects in CESM based on the first 5 years of a coupled simulation (Figure 5a) are similar in sign (~20 to 30ppbv) but also not significant as for GATOR-GCMOM.

3.2.2 Effects on ozone with CAM5 using fixed meteorology fields

CAM5 was also run with fixed meteorology fields for 2050 (based on meteorology from free running simulations). The NOx-induced changes in tropospheric ozone are complicated by a short-term increase in O_3 concentrations associated with a positive forcing, and a long-term reduction of O_3 concentrations tied to the aviation induced methane decrease. The long term-reduction is associated with negative forcing (Stevenson et al., 2004). The short-term O_3 forcing is one of the major contributors to the overall aviation forcing and dominates the net O_3 forcing (Lee et al., 2009). Since CH_4 mixing ratios at the boundary layer are fixed, the calculated changes in O_3 concentration are the short-term changes.

Radiative forcing effects from these aviation perturbations are illustrated in Table 2 for each of the scenarios. Enhanced upper tropospheric and lower stratospheric ozone (O_3), termed "short-term O_3 forcing", due to nitrogen oxide (NOx) emissions contributes ~60mWm^{-2} of radiative forcing. Changes to the methane lifetime induced by aviation are estimated to induce a radiative forcing of -25mWm^{-2} in 2050.

3.2.3 Alternative fuel effects on ozone

Figure 3A and B show the effect of using an alternative fuel on ozone. Both CESM and GATOR-GCMOM indicate very small effects on ozone by switching from traditional fuel to alternative fuel with reduced BC and no sulfur. Values are not significant. The aviation NOx-induced effect on ozone in the presence of sulfur and BC appears to be the same as for standard fuel.

3.4 Temperature Effects

GATOR-GCMOM and CESM were run coupled to an active ocean to estimate the temperature and surface temperature response to aviation emissions in 2050. The major motivation for running fully coupled climate simulations with GATOR-GCMOM and CESM is to assess the effects of aviation radiative forcing due to chemistry, contrails and aerosols on surface climate.

3.4.1 Surface Temperature changes

Surface temperature changes are shown in Figure 4 for GATOR-GCMOM for the first 5 years for the effects of baseline emissions (Figure 4A) and the alternative fuel Scenario 2B (Figure 4B): very few of these differences pass a significance test in any coherent way. Similar results are seen for CESM simulations in the first 5 years (not shown). CESM is still adjusting to the forcing. CESM results over the years 31-50 for baseline (Figure 4C) and the effects of an alternative fuel (Figure 4D) show more significant patterns. Jacobson et al (2012) found with similar 20-year GATOR-GCMOM experiments that 16% of the planet with 'significant' temperature changes (>95% confidence level), including warming in the Arctic after a 20 year coupled experiment with present day aviation emission. In Figure 4 (and in Jacobson et al 2010), regions of significant surface temperature change are not precisely co-located with the largest forcing. For baseline emissions, net cooling from aviation sulfate in CESM causes significant Northern Hemisphere mid-latitude cooling (Figure 4C), consistent with the effect of the net negative radiative forcing for the baseline scenario in Figure 1b. The removal of sulfur in the Alternative Fuel Scenario 2 causes a reversal of this pattern (significant warming), associated with an increase in net radiative forcing (Figure 1B: scenario 1 to scenario 2).

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We have also examined CESM initialized with a similar ocean structure to GATOR-GCMOM. The ocean initial condition is a uniform temperature scaling with depth. The CESM simulation with such ocean initial conditions is not in balance, and over the course of the 5 years analyzed is still adjusting. This simulation was conducted to better compare with GATOR-GCMOM, and uses the same baseline emission scenario as GATOR-GCMOM. It highlights the transient nature of signals in the first 5 years of simulation. A case could be made that it is sufficient to look at the perturbation to the transient response between the two simulations, but the transient and longer term response seem different in CESM simulations because of the out of balance initial conditions from GATOR-GCMOM. Even after 50 years with a small perturbation any the dis-equilibrium is smaller than the internal variability of the climate system (i.e., not significantly different than zero change).

3.4.2 Vertical Temperature structure

Figure 2 illustrates the impact of baseline aviation emissions on zonal mean temperature in GATOR-GCMOM (Figure 2C) and CESM (Figure 2D). Different time periods are shown: first 5 years for GATOR-GCMOM and years 31-50 for CESM. CESM is similar to GATOR-GCMOM over the first 5 years (Figure 5). Over the first 5 years there is warming in the tropics and warming at flight levels, but cooling at higher altitudes and high latitudes. However, very little of this signal is significant given the variability in a 5 year simulation. For CESM some of the tropical warming and warming in the N. Hemisphere at flight altitudes is significant over the first 20 years of simulation (Figure 5) but this becomes a significant cooling over the last 20 years of a 50 year simulation due to the radiative cooling from increased aviation sulfur, as noted below (Gettelman and Chen 2013).

In GATOR-GCMOM, BC is a strong contributor to the UTLS warming, and the reduction of BC in the Alt fuel scenario (Figure 3C) reduces warming relative to the baseline, noted below. The same is seen in CESM in the first 5 years of simulation (Figure 6). The warming due to BC (and cooling from its removal) occurs due to two factors. First, there is enhanced BC absorption upon coating by sulfate. Second, by enhanced BC absorption between contrail ice particles (Jacobson et al., 2013). To obtain the full effect of enhanced absorption, it is necessary to treat the hygroscopic growth of BC-coated aerosol particles at the relative humidity of the cloud rather than at the relative humidity of the clear sky. In addition, over snow-covered, ice-covered, and cloud surfaces, BC absorbs not only solar radiation going down but also reflected upward radiation. As such, it impacts over reflective surfaces exceed those over darker-surfaces.

Contrails themselves contribute to upper-tropospheric warming at sub-polar latitudes, in part due to enhanced solar absorption due to their black carbon inclusions within contrail particles. Reducing BC (not shown) alone results in less upper tropospheric warming compared with low-BC and low-sulfur fuel. The combination of aircraft SOx, NOx, CO, and HCs (and their effects on O_3) has much less of an impact on temperatures than does BC alone. Thus most of the UTLS warming

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in GATOR-GCMOM (Figure 2C) appears to be due to BC. BC as pure aerosol warms the air by direct absorption of solar radiation. When BC is emitted from aircraft however it is coated by organic material, sulfate, and water. Coating increases BC absorption due to the optical focusing effect (Jacobson et al., 2013).

3.4.3 Alternative Fuel Effects

The reduction of BC in the alternative fuel scenario reduces the flight level warming in GATOR-GCMOM (Figure 3C) and in CESM (Figure 3D and Figure 6), though neither is significant over a 5-year period. Over the last 20 years of a 50-year simulation (Figure 3D), the reduction of Sulfur in CESM alternative fuel Scenario 2 results in a significant warming near the surface in the N. Hemisphere as contrail radiative forcing dominates and the cooling seen in Figure 2D is reversed when sulfur is removed in the alternative fuel scenario. This is seen globally in Figure 1, and in the vertical temperature structure of Figure 3. The evolution of the temperature structure in the Alt Fuel scenario for CESM (Figure 6) highlights that the vertical structure of temperature changes quickly due to local radiative forcing, but then evolves in response to surface temperature effects. The BC effect is a local warming at altitude, while the brightening of clouds due to aviation sulfur affects the surface, which takes years to evolve. How this happens is best understood by looking in detail at the pattern of surface temperature changes.

4. Discussion

The major mechanisms by which aviation affects temperature differ between the two models. For GATOR-GCMOM, absorption due to aviation BC is important. For CESM, over land, contrail cirrus dominate the radiative forcing, while over oceans, this is negated by cooling due to sulfate brightening of low level liquid clouds. However, the initial responses of the models over the first 5 years of simulation are broadly similar. Below we attempt to understand and assess these differences.

4.1 Sulfur

Aviation sulfur affects climate in two ways. (1) Sulfate nucleates liquid cloud drops by deliquescence and activation of haze particles. (2) Sulfuric acid in the hot exhaust will condense onto BC particles to enhance the number concentration of contrail particles. Nucleation, (1), is unique to the co-emission of sulfur and BC (i.e., from aviation). Both models treat nucleation. GATOR-GCMOM treats sulfur-BC mixtures whereas CESM assumes BC and sulfate are externally mixed in the exhaust. In addition, sulfate coating of BC changes the absorption and ice nucleating properties of the BC aerosols in GATOR-GCMOM with both aviation and background emission. Aircraft aerosol sulfate is a small fraction of total aircraft-emitted sulfur, as much of the sulfur in both models manifests as SO_2, but that small amount of sulfate is a relatively large fraction of total emitted aerosol mass.

Possibly the main reason for the difference in sulfate cooling due to aviation in CESM (Figure 1) versus a small warming in GATOR-GCMOM (discussion in section 3.4.1) is that CESM predicts a much lower background (non-aviation) aerosol number concentration for particles >100nm, only 6% that of GATOR-GCMOM, so changes in emissions due to aircraft, have a greater impact on cloud indirect effects in CESM. CESM has larger (10%) perturbations to the number of Cloud Condensation Nuclei (CCN) in 2050 in N. Hemisphere mid-latitudes due to aviation. These perturbations contribute to low-cloud brightening and cooling. Total Aerosol Optical Depth (AOD) levels (not shown) are much closer between CESM and GATOR-GCMOM because of similar numbers of small particles <100nm. In GATOR-GCMOM the higher background particle numbers and lower percent perturbation mean that aviation sulfur has less impact on clouds.

Previous work from Righi et al (2013) treats aerosols similarly to CESM, and finds similar cooling effects to CESM. However, Righi et al. (2013) did not treat cloud absorption effects of BC aerosols emitted by aircraft, and did not examine climate response, only radiative forcing.

The impact of sulfur aerosols indicates that significant changes to aviation sulfur emissions will have large effect on the climate response to aviation emissions.

4.1.2 BC (Soot)

GATOR-GCMOM has a large impact from the effects of enhanced BC absorption at altitude, due to (a) direct effects of sulfur coated BC, (b) cloud absorption effects of BC in contrail ice, (c) semi-direct effects, and (d) indirect effects on contrail ice number. GATOR-GCMOM treats these effects, including the coating of BC particles by sulfate, ammonium, water, and other aerosol constituents, enhancing cloud absorption effects of BC aerosols. At the high super-saturations seen in aircraft exhaust, all BC particles should activate to small liquid drops that eventually freeze to form ice crystals in the plume. These may then coalesce into larger drops.

The Modal Aerosol Model (MAM) in CESM does mix BC and sulfate aerosol and simulate enhanced absorption due to internally mixed aerosol BC, but MAM in CESM does not treat enhanced absorption by contrail particles containing BC. CESM does not treat enhanced absorption by aerosol BC between non-contrail cloud particles at the relative humidity of the cloud or within cloud particles. CESM does not assume an initial particle number based on BC, but rather an 'aged' or coagulated particle number based on coagulated particles (10 microns in size). The BC aerosol absorption enhancement due to mixing of BC aerosol with sulfate is included in CESM, but there are significant differences in the treatment of particle sizes and optical properties between the grid scale model of CESM and the plume model in GATOR-GCMOM. However, we do see similar initial warming in CESM as in GATOR-GCMOM in the baseline scenario (Figure 2 and Figure 5) before effects of sulfur cooling in CESM alter surface temperature.

We tested any differences in the size distribution of BC between CESM and GATOR-GCMOM, altering CESM simulations to reflect smaller BC size and higher numbers. CESM makes no distinction between background BC and aviation BC, and CESM assumes 0.1% efficiency of ice nucleation for all BC particles (Gettelman et al., 2010). The plume processing that occurs in GATOR-GCMOM first allows for coagulation that reduces BC particle number significantly. Then, nearly 100% of the resulting BC particles can activate to form contrail particles in the highly-supersaturated contrail plume that exists immediately behind an aircraft (Jacobson et al., 2011).

To explore the effect of this difference, CESM simulations were performed where the contrail ice particle number concentration was set to the aviation soot number, both in a specified dynamics (to get radiative forcing) and coupled simulation. This results in much higher ice crystal numbers in contrails in CESM, more similar to what GATOR-GCMOM would produce in a contrail. In this test, the positive cloud forcing in SD simulations from contrail cirrus in CESM increases by a factor of 4 and there is a net warming that is more similar to GATOR-GCMOM in magnitude. The warming is large enough in this simulation to reduce contrail formation due to the warming (burnoff) as in GATOR-GCMOM.

From this analysis, it appears that the increased contrail number due to BC aerosols and absorption creates a larger warming in GATOR-GCMOM. Similar effects can be produced in CESM with similar assumptions of higher contrail numbers from activation on BC and limited coagulation. Previous studies showed similar effects. For example, Wong et al. (2013) found using laboratory experiments and modeling studies that soot was internally mixed in contrail ice particles and Liou et al. (2013) found a resulting increase in cloud heating rate due to these internal mixtures.

Thus it appears that the BC soot aerosol particles emitted by aircraft increase the number of contrail ice crystals, increasing the optical thickness, for a net warming (as the enhanced longwave absorption dominates over the enhanced shortwave reflectance). This seems in contrast to significant cooling from soot effects on cirrus clouds noted by Zhou and Penner (2014).

The effects of soot can significantly change the climate impact of aviation. To further constrain the BC soot effects for climate purposes, it would be necessary to characterize better the near term evolution of ice particles and try to constrain better the particle size distributions in contrails. Further analysis of existing observations of such size distributions against the model size distributions could be conducted, but that for the moment is beyond the scope of this work.

4.1.3 Ozone

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Both models show ozone concentration enhancement in the upper troposphere and lower stratosphere (9-12km) in the N. Hemisphere (e.g., Figure 2A from GATOR-GCMOM) from the effects of aviation emissions. CESM shows similar effects for simulations run as a CTM, without differences in meteorology or temperature. Neither model shows significant effects from the alternative fuels scenario on the changes in ozone concentrations (Figure 3A and 3B). However, these effects are seen in the CESM uncoupled SD experiments with a lower noise threshold (Table 2). Ozone changes are consistent with NOx and NOy changes in CESM. Increases in ozone concentrations are consistent with previous work assessed in Brasseur et al (2015).

4.2 Surface Temperature

Both GATOR-GCMOM and CESM performed 2050 simulations of surface temperature changes. The positive radiative forcing in GATOR-GCMOM contributes to warming in the simulations, but it is not significant in a five-year simulation in either model. Most of the effect is in the atmosphere and the surface takes time to react. In general, surface temperature effects of aviation emissions in 2050 are barely statistically significant in most regions and not significant globally. Similar results were found for present day emissions by Jacobson et al. (2012) with 20-year GATOR-GCMOM experiments. The cooling from indirect effects of aviation sulfur in CESM results in a global average mean temperature change of -0.11K after 50 years (Figure 4C), but this is not significantly different from zero. This means that the short term aviation signal from contrails and aerosols, even at 5-8 times current levels in 2050, does not have a significant globally-averaged surface temperature signal.

Regional patterns of temperature change in Figure 4 are broadly correlated with short term aviation radiative forcing. For CESM simulations with strong cooling from sulfate, there is cooling over the N. Atlantic, particularly in regions where deep water is forming in the ocean (Figure 4C). There is a reduction of this cooling (warming) if effect of sulfur is removed with an alternative fuel (Figure 4D). Note that the warming and cooling in the coupled system occur not necessarily where the largest forcing is (over N. America and Europe), but where the climate system is most sensitive to perturbations (the sea ice edge and the region of deep water formation). Also, the climate system takes time to react to the forcing at the surface, particularly over the ocean.

5. Conclusions

This study has analyzed the climate effects of 2050 aviation emissions using the same 2050 emissions scenarios with two very different climate-chemistry modeling systems, Simulations indicate significant growth in the climate impact of aviation that occurs with projected increases in fuel use. Fuel use increases by a factor of 3 to 5 relative to 2006, while radiative

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forcing from contrails increases 12 to 90 mWm^{-2} (7 times 2006 forcing). The baseline scenario (the high end of this range) assumes no technology improvement and is likely an unrealistic upper bound. It is shown here for comparison purposes. Regional radiative forcing for 2050 is concentrated over regions of high emissions, either directly over high traffic land regions of Asia, N. America and W. Europe, or over oceanic flight corridors of the N. Pacific and N. Atlantic. Regional temperature change happens where the climate system is most sensitive near regions of large forcing, but not always coincident with largest forcing.

Simulations in both model systems indicate that "non-linear" effects of aviation emissions, chiefly through interactions with aerosol particles, can be larger than the effects of aviation CO_2 or aviation H_2O emissions that cause contrails (Figure 1). The main particle emissions affecting radiative forcing are sulfate and BC soot.

Sulfur is emitted primarily as a gas and subsequently evolves in the atmosphere into sulfate aerosols that can interact with liquid clouds. The effect of this process will be net cooling, but mostly over oceans. Because CESM predicts lower background aerosol concentrations of larger aerosol particles, changes in emissions due to aircraft increase aerosol loadings by a larger fraction in CESM resulting in a greater impact on indirect effects in CESM. As such, CESM predicts more cloud-enhanced cooling than GATOR-GCMOM of as much as -150 mWm^{-2} in CESM in 2050 (Figure 1C).

GATOR-GCMOM indicates that BC, coated by sulfate and efficiently activating to form contrail particles increases contrail absorption (heating), while CESM does not simulate sulfate coating BC in contrail plumes to increase activation rates. However, there is still warming at altitude in CESM either from BC or from contrails. When large BC numbers are assumed to efficiently create contrail particles in CESM, there is better agreement in results between the two models. Thus, BC emissions appear in both models, if treated similarly, to cause increases in contrail absorption (net heating).

As a result of these sulfur and BC effects, both model simulations indicate that the use of alternative fuels that substantially reduce sulfur and BC emissions from aviation may significantly alter the future climate impact of aviation. These effects are large in both models examined, but depend on the nature of the processes represented (BC enhanced absorption in GATOR-GCMOM) and the background state (lower sulfate in CESM). The magnitude of potential positive forcing from BC effects still needs to be determined and better constrained.

The different non-linear effects indicate a very important role for the different emission scenarios. Reduced fuel burn (Scenario 1) reduces impacts across all different aviation components. Alternative fuels with lower BC and no sulfur (Scenario 2 and 2B) significantly reduce non-linear effects of aerosol sulfur and BC while not affecting contrail formation very much. The result would shift the balance of aviation radiative forcing effects but with reductions of positive forcing for

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BC reductions, and reduced negative forcing for sulfur reductions. There are still large uncertainties in this finding that need to be better assessed by more fully analyzing how the models treat aviation BC soot and sulfate aerosols.

Ozone responses in 2050 to aviation emissions indicate increases in ozone in flight corridors of 20-60ppb. These effects from aviation may be large (particularly in otherwise pristine regions), depending on the future background emissions. Short term O_3 forcing is estimated at ~60mWm^{-2}, and changes to the methane forcing induced by aviation-caused methane lifetime changes are estimated at -25mWm^{-2} in 2050 (based on differences with and without aviation).

The global surface temperature effects of aviation in 2050 are small, and not significantly different than zero in almost all regions. Effects in the Arctic seen in GATOR-GCMOM (Jacobson et al. 2013) are also seen in some CESM simulations over short periods of time, but not in longer CESM coupled simulations. This means that some of the transient differences may be a product of the slightly different evolution of feedbacks in the transient GATOR-GCMOM and CESM simulations. Effects in CESM are seen over longer periods (Figure 4C and 4D) in response to large forcings. Arctic effects are seen in CESM only in simulations set up with an ocean state out of balance. However, the effects of temperature were not examined with CESM with background aerosol levels similar to those in GATOR-GCMOM. Thus high latitude effects, particularly surface temperature responses, should be studied more, but will require long equilibrated simulations with coupled models.

Regional radiative forcing does NOT translate into regional surface temperature responses even when regional forcing reaches 2Wm^{-2}. The heat budget of middle and high latitudes is not controlled locally, but has a large transport component. Over regions of the largest positive aviation radiative forcing over continents, temperature may be more due to advection than local radiative fluxes. Persistent short term regional forcing does impact the climate system where the system is most sensitive (i.e., regions of the sea ice edge and deep water formation) rather than where forcing is largest. The impact on the surface takes time to develop after radiative forcing is integrated for many years. Thus, short-lived and non-uniform radiative forcing (e.g., from aviation aerosols and contrail effects) may have a climate response per unit of forcing very different from long-lived and uniform forcing (such as from aviation CO_2). Local effects on temperature may not be correlated with local radiative forcing. Long coupled simulations are needed to be able to observe these effects, even for 2050 emissions whose forcing is several times that of 2006.

The diurnal timing of contrail effects of aviation may mean that there are some regional effects not treated here, though diurnal cycle effects (Chen and Gettelman, 2013) are smaller than uncertainties in the model formulations or emissions.

Thus non-linear effects of soot and sulfur from aviation will strongly affect the overall climate impact of aviation in 2050. The pathways indicated by the two modeling systems are similar. These results strongly indicate that use of alternative fuels that significantly reduce sulfur and BC emissions from aviation will significantly alter the future climate impacts of aviation:

sulfur is a strong cooling effect through indirect effects on clouds, and soot heats the upper troposphere in the simulations with some net surface warming. Both these effects would be mitigated with alternative fuels. The effects are uncertain, and further comparisons between models and observations of contrail aerosols would be valuable and important to better constrain these effects.

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TABLES

Table 1: Description of Emissions Scenarios used in this work.

Scenario List	
Baseline	Baseline Fuel Burn (technology improvements not included)
Scenario 1	Reduced Fuel Burn (includes assumed technology improvements)
Scenario 2	'Alternative Fuel'; Reduced Fuel Burn, no Sulfur, 50% BC
Scenario 2B	GATOR-GCMOM Baseline 'Alt Fuel' Scenario
Scenario 3	Scenario 2 with 5% increased H_2O

Table 2 Comparison of aviation emissions component specific RF (mW/m^2) for 2006 and 2050 baseline and alternative scenarios (From Brasseur et al., 2015, Table 4) with new results from this study added in bold.

Scenario	**Fuel burn (Tg)**	**NO_x (Tg N)**	**O_3-S**	**CH_4**	**Long Term O_3**	**Water Vapor**	**Contrails**	**Aerosols**
			UIUC CAM5*	**UIUC CAM5***	**UIUC CAM5**	**UIUC CAM5**	**NCAR CAM5**	**NCAR CAM5**
2006 -Base	188.1	0.812	36.5	-12.3	-4.5	-2.6	17	-38
2050-Base	902.8	3.950	143	-59.7	-20.3	-12.5	**83**	**-160**
2050-S1	514.4	1.570	70.5	-28.3	-9.4	-5.9	**72**	**-107**
2050-S2	514.4	1.570	**58.5**	**-25.6**	**-8.8**	**-5.4**	**72**	**0**

Figures

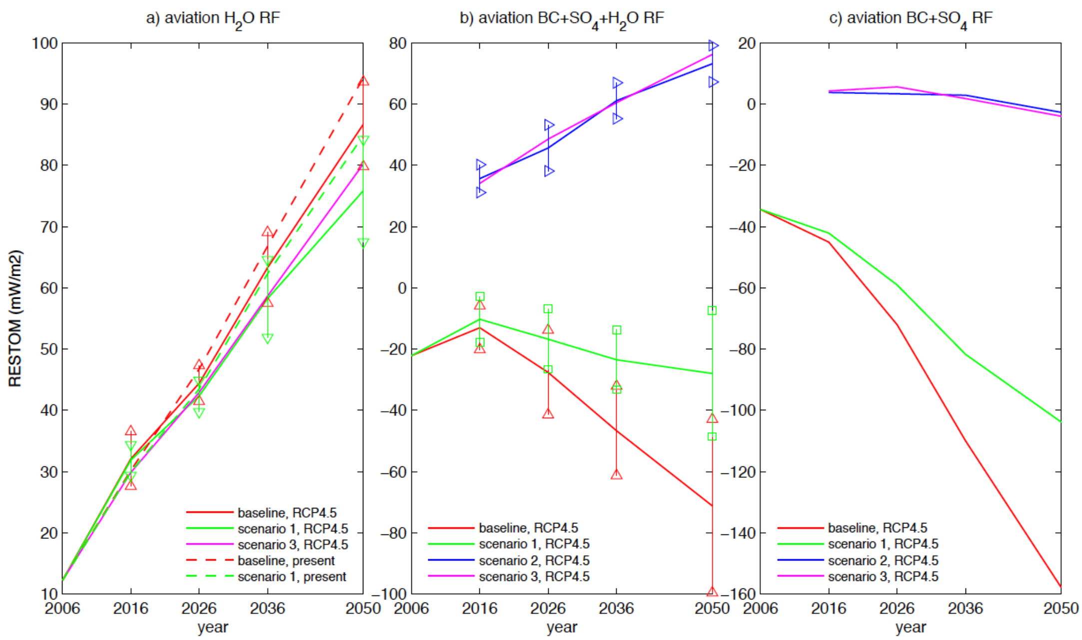

Figure 1: Radiative forcing in CESM from (A) H_2O emissions alone (contrail cirrus), (B) H_2O +BC + SO_4 emissions and (C) The residual effect of BC and SO_4 estimated from (B) – (A). Solid lines are for the RCP 4.5 scenario with different meteorology each year; dashed lines in panel (A) are using 2006 meteorology. Since scenario 1 and scenario 2 have the same H_2O emissions, they have the same RF in (A). The baseline aviation scenario is in red, scenario 1 is green, scenario 2 is blue and scenario 3 is purple.

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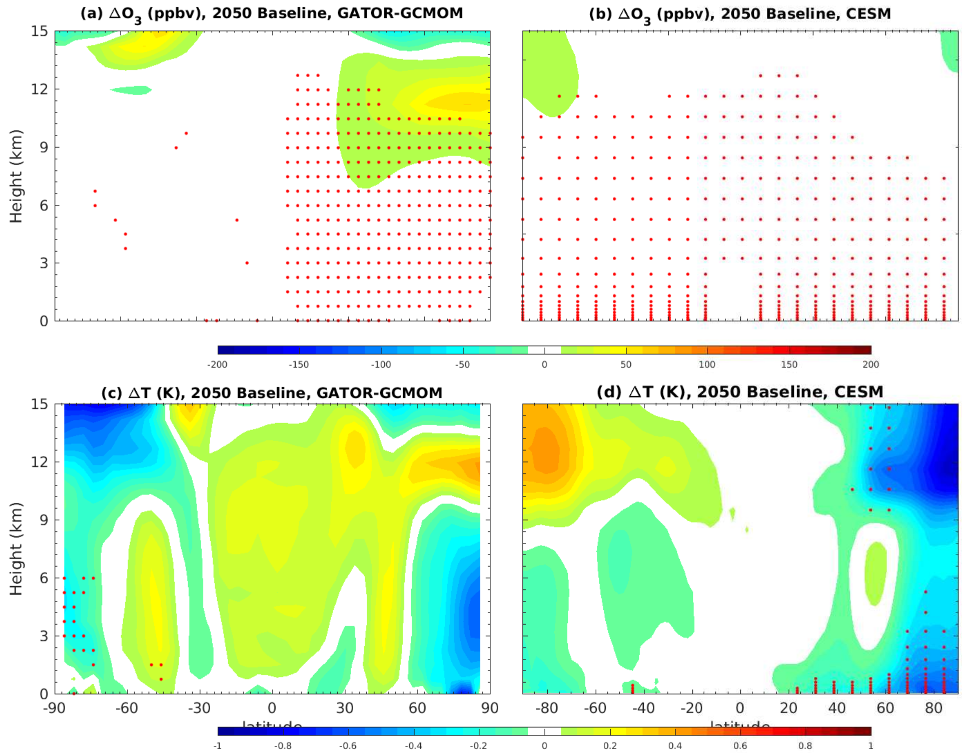

Figure 2: Zonal mean change in (A,B) ozone and (C,D) temperature due to baseline 2050 aviation emissions (v. no aviation emissions). Panels A,C for GATOR-GCMOM and B,D CESM. Red dots show regions of statistically significant perturbations. Based on first 5 years of simulations for GATOR-GCMOM, and years 31-50 for CESM.

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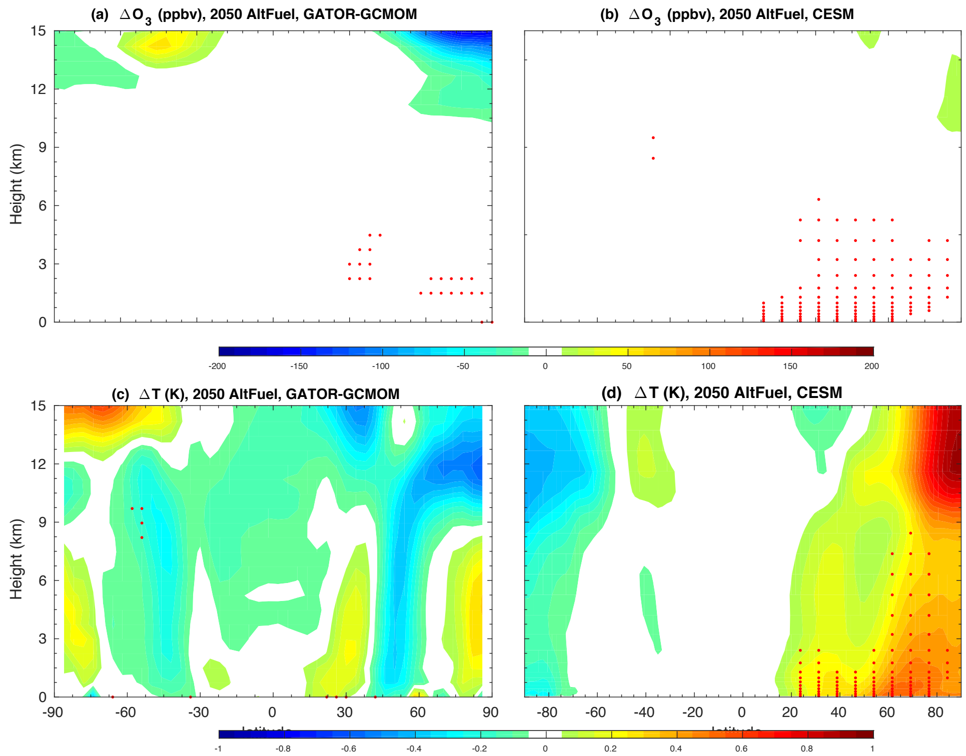

Figure 3: Zonal mean change in (A,B) ozone and (C,D) temperature due to Alternative Fuel 2050 aviation emissions (v. 2050 aviation emissions). Panels A,C for GATOR-GCMOM and B,D CESM. Red dots show regions of statistically significant perturbations. Based on first 5 years of simulations for GATOR-GCMOM, and years 31-50 for CESM.

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Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 April 2017

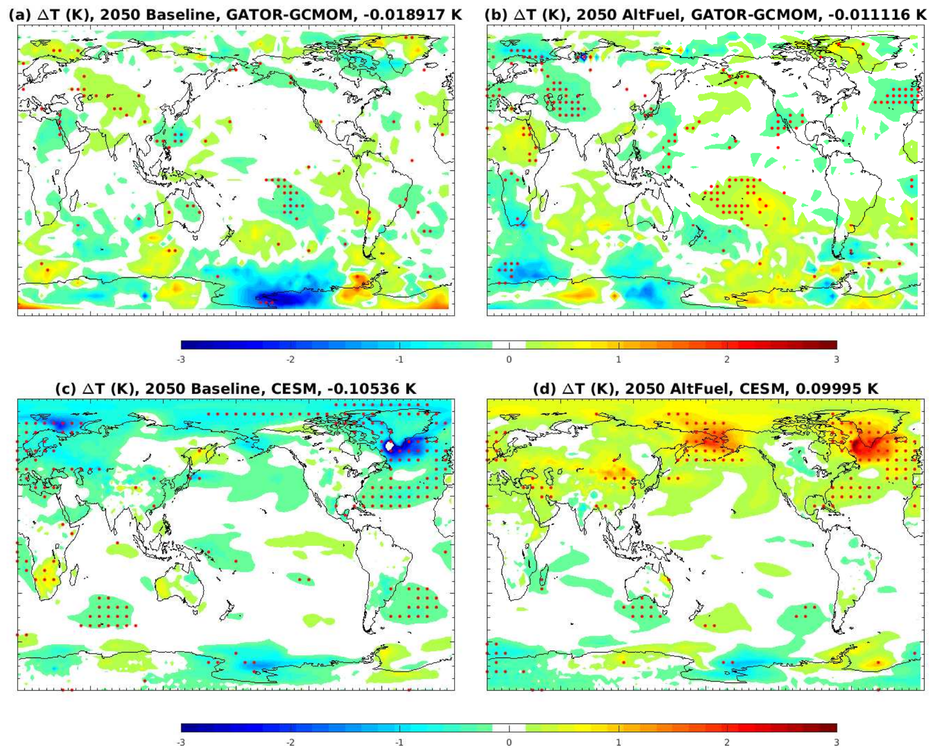

Figure 4: Surface temperature changes in (A, B) GATOR_GCMOM and (C, D) 50 year CESM coupled simulations. A and C are for baseline emissions (v. no emissions) and B and D are for an alternative fuel scenario. Red dots show regions of statistically significant perturbations. Red dots show regions of statistically significant perturbations.

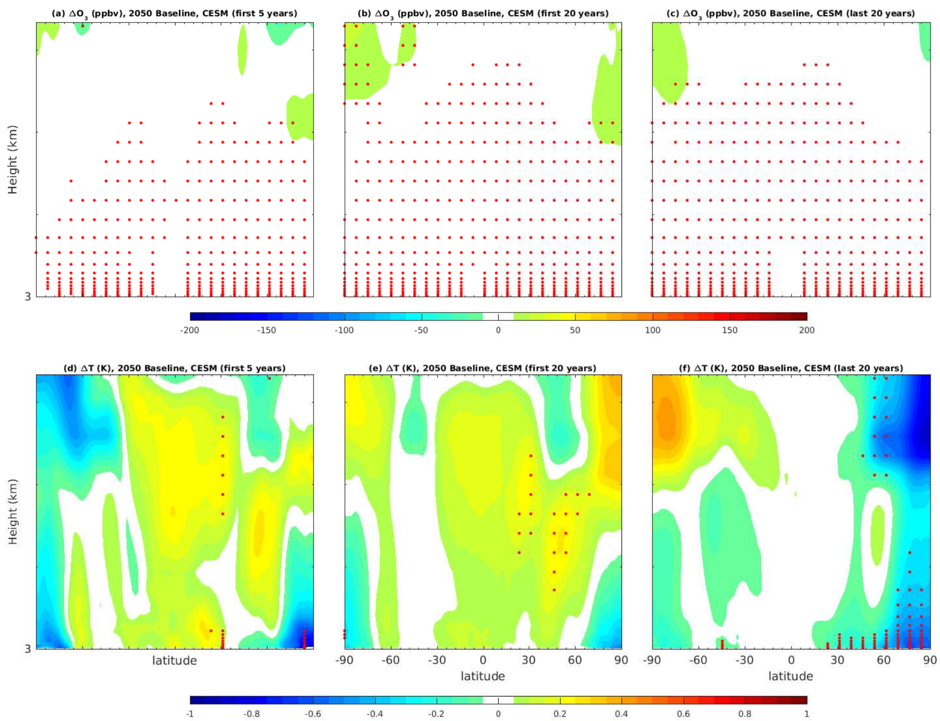

Figure 5: CESM simulations for different time periods. Baseline – No Aviation. (A,B,C) dO3, (D,E,F) dTs. (A,D) First 5 years, (B,E) First 20 years, (C,F) Years 31-50. Red dots show regions of statistically significant perturbations.

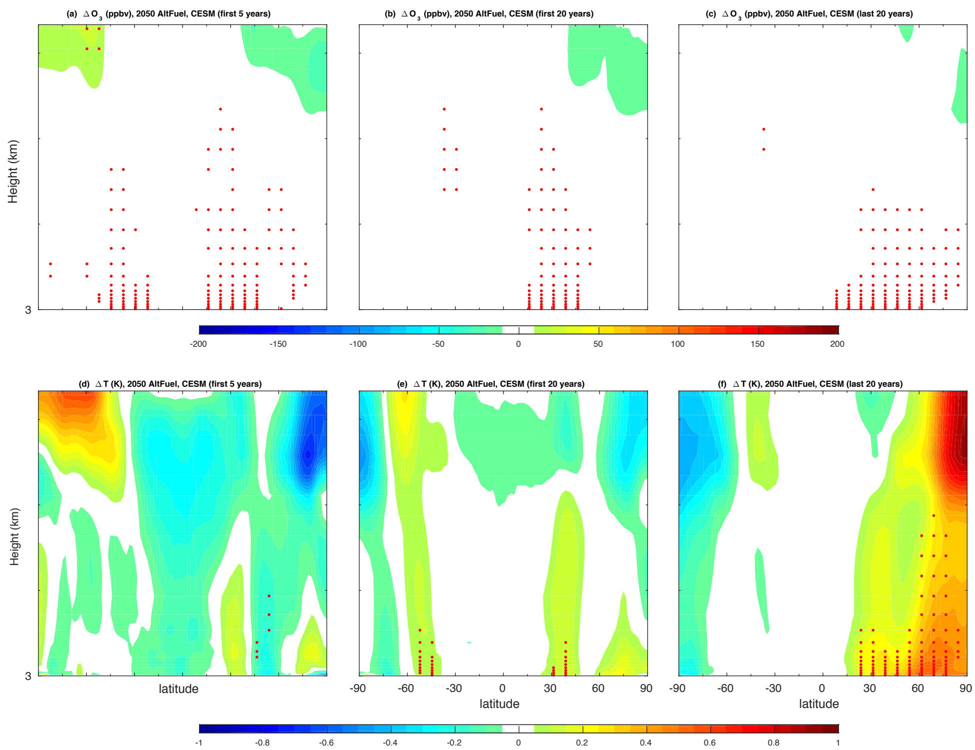

Figure 6: CESM simulations for different time periods. AltFuel – Regular fuel (Scenario 2 – Scenario 1) (A,B,C) dO3, (D,E,F) dTs. (A,D) First 5 years, (B,E) First 20 years, (C,F) Years 31-50. Red dots show regions of statistically significant perturbations.