The climate impact of aviation aerosols

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[1] A comprehensive general circulation model (GCM) is used to estimate the climate impact of aviation emissions of black carbon (BC) and sulfate (SO_4) aerosols. Aviation BC is found not to exert significant radiative forcing impacts, when BC nucleating efficiencies in line with observations are used. Sulfate emissions from aircraft are found to alter liquid clouds at altitudes below emission ($\sim$ 200 hPa); contributing to shortwave cloud brightening through enhanced liquid water path and drop number concentration in major flight corridors, particularly in the N. Atlantic. Global averaged sulfate direct and indirect effects on liquid clouds of 46 mWm^{-2} are larger than the warming effect of aviation induced cloudiness of 16 mWm^{-2}. The net result of including contrail cirrus and aerosol effects is a global averaged cooling of -21 ± 11 mWm^{-2}. These aerosol forcings should be considered with contrails in evaluating the total global impact of aviation on climate. **Citation:** Gettelman, A., and C. Chen (2013), The climate impact of aviation aerosols, *Geophys. Res. Lett.*, *40*, doi:10.1002/grl.50520.

1. Introduction

[2] Aviation fossil fuel emissions are a small ($\sim$2%) but growing component of anthropogenic perturbations to the atmosphere [*IPCC*, 1999; *Lee et al.*, 2009]. In addition to emissions of CO_2, aircraft emit water vapor, forming contrails. Contrails heat the planet (as high optically thin clouds have larger longwave heating than shortwave cooling effects). Aircraft emissions of nitrogen oxides cause perturbations to ozone (increases and decreases) and decreases in methane, which both have radiative effects [*Lee et al.*, 2009]. Aircraft also emit aerosols or their precursor gases, chiefly sulfate (SO_4) and Black Carbon (BC, also called soot), that directly reflect shortwave solar radiation (for SO_4) and absorb shortwave solar and longwave terrestrial radiation (for BC). These aerosols may also alter cloud and ice nuclei and resulting drop and crystal concentrations, inducing an indirect effect on climate by changing cloud radiative effects [*Twomey*, 1977].

[3] *Lee et al.* [2009] found aviation CO_2 radiative forcing in 2005 was 30 mWm^{-2} (1000 mWm^{-2} = 1 Wm^{-2}). *Lee et al.* [2009] estimated direct effects of SO_4 to be –5 mWm^{-2} and 4 mWm^{-2} for BC. For linear contrail radiative forcing, *Lee et al.* [2009] reported 12 mWm^{-2}. *Burkhardt and Kärcher* [2011] reported integrated radiative forcing of contrails (contrail-cirrus) of 35 mWm^{-2}.

[4] There have been fewer studies of the influence of aviation aerosols on clouds (indirect effects). *Hendricks et al*. [2005] and *Hendricks et al.* [2011] found significant increases in ice crystal number due to aviation BC if the aerosols are efficient ice nuclei. *Penner et al.* [2009] using the aviation emissions inventory in *Lee et al.* [2009] in a chemical transport model found large cooling effects from the indirect effects of aircraft BC emissions of –161 and –124 mWm^{-2} due to increasing ice number concentrations in regions of emissions (but decreases elsewhere), depending on ice nucleation parameterization and assuming high efficiency of BC as heterogenous ice nuclei.

[5] We use a detailed general circulation model (GCM) that represents key physical processes (ice supersaturation and ice nucleation on aerosols) with a new aviation emissions data set to simulate the integrated effect of aviation aerosol emissions on clouds and climate. Models and experiments are described in section 2, results are presented in section 3, and conclusions are in section 4.

2. Models and Experiments

2.1. CAM5 Description

[6] We use the Community Atmosphere Model version 5 (CAM5) from the National Center for Atmospheric Research (NCAR). CAM5 has a two moment cloud microphysics scheme [*Morrison and Gettelman*, 2008], coupled to a Modal Aerosol Model with three modes [*Liu et al.*, 2012]. The model allows ice supersaturation and links ice cloud particles consistently to aerosols and radiation through ice nucleation [*Liu et al.*, 2007; *Gettelman et al.*, 2010]. We include 0.1% of the black carbon (BC) as an efficient ice nucleus for heterogenous freezing (with similar properties to mineral dust) in the base case, consistent with BC found in ice residues [*DeMott et al.*, 2009, 2010].

[7] We perform simulations with Aviation Environmental Design Tool (AEDT) aviation emissions [*Barrett et al.*, 2010] as described by *Chen et al.* [2012]. Simulations are detailed in Table 1. We focus on water vapor (H_2O), SO_4, and BC. We do not simulate the effects of Aviation NO_x on ozone or methane. As in *Chen and Gettelman* [2013], H_2O emissions are evaluated for whether environmental conditions support persistent contrail formation. If so, H_2O is injected as cloud ice, with specified fraction and microphysical properties. In the H2O and "ALL" simulation, contrail ice (and/or vapor from aircraft) is treated like any other ice cloud in CAM5 after being initialized as a contrail [*Chen et al*., 2012] and interacts with the model physics, radiation, and dynamics.

[8] Aerosols do not impact the initial contrail ice microphysics, but subsequently affect nucleation. For the sulfate (SO4) and black carbon (BC) simulations, we add SO_4 and BC mass and number to the aerosol model modes. A

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Table 1. Description of Simulations Used in This Study

Name	Description
Base	CAM5 base code: no aviation aerosol or contrail emissions
SO4 (8, 14, 21 nm)	CAM5 + aviation SO_4 emissions
BC-direct (11, 38, 79 nm)	CAM5 + aviation BC emissions, no BC ice nucleation
BC 2% (11, 38, 79 nm)	CAM5 + aviation BC emissions, 2% efficiency
BC 0.1% (38 nm)	CAM5 + aviation BC emissions, 0.1% efficiency
H2O	CAM5 + aviation H_2O emissions & contrails
ALL	CAM5 + aviation emissions of H_2O (Contrails), BC (38 nm) & SO_4 (14 nm)

log-normal size distribution with geometric standard deviation of 1.6 is used [*Barrett et al.*, 2010]. SO_2 does not impact ice nucleation directly, but evolves in the aerosol model as described by *Liu et al.* [2012]. We use geometric diameters of 14 nm for SO_4 and 38 nm for BC [*Barrett et al.*, 2010] to calculate initial aviation aerosol number. The aerosol model then creates internal mixtures. We also evaluate extremes of the size ranges in *Barrett et al.* [2010] for SO_4 (8 and 21 nm) and BC (11 and 79 nm). With constant mass, smaller sizes result in higher number concentrations. We also evaluate different efficiencies for BC nucleation: either 0% (direct effects of aerosols only), 0.1% (base case), and 2% (high efficiency).

[9] CAM5 simulations are performed using specified dynamics and monthly averaged emissions. Temperatures and winds are prescribed based on 1 year of output from a free running CAM simulation with climatological SSTs from 1979–2000. Cloud parameterizations and water vapor sources and sinks are allowed to operate as normal. H_2O is conserved and advected. For a 5 year simulation, the 95% statistical significance level for detection of a global top of atmosphere radiative perturbation is $\sim$10 mWm^{-2}. Regionally, the significance level is between 100 and 200 mWm^{-2} over the North Atlantic [*Chen and Gettelman*, 2013]. Statistical significance is included in the regional results. Simulations are 5 years in length, and the same meteorology is repeated each year. For the combined simulation ALL, we run four ensemble members with CAM5 temperatures and winds from a different simulation year and report ensemble mean and two standard deviation (σ) meteorological uncertainty in Table 2.

[10] Uncertainties in these estimates of aviation impacts come from several sources: (1) meteorological "noise," sensitivity to (2) aviation and (3) background aerosol mass and number, and (4) uncertainties in the contrail parameterization. In this work, we assess (1) with ensembles, and test for significance using this variability. We assess (2) by altering the size distribution of aviation aerosols (which affects number concentrations). *Barrett et al.* [2010] report uncertainties in mass emissions factors of 10% for H_2O, 30% for SO_4 and 100% for BC. Uncertainty in the fuel mass burned is the subject of ongoing work on future emissions projections. We do not assess (3), but note that the model has been evaluated extensively against observations and has a realistic background state [*Gettelman et al.*, 2010; *Chen et al.*, 2012]. The contrail parameterization (4) has meteorological uncertainty of about $\pm$30% [*Chen and Gettelman*, 2013] and parametric uncertainties for the radiative forcing based on assumptions about initial crystal size and mass uptake estimated to be a factor of 2 by *Chen and Gettelman* [2013].

[11] Aerosol direct and indirect effects are assessed by differences between simulations with and without aviation aerosols. Combined effects are given by the change in Top of Atmosphere (TOA) flux (ΔR). Indirect effects are estimated by the change in Cloud Radiative Effects (ΔCRE), the sum of long wave (ΔCRE_{LW}) and shortwave (ΔCRE_{SW}) components. Direct effects are diagnosed by the change in TOA SW clear sky flux (ΔR_{SWclr}), or the residual of the total and indirect effects (ΔR - ΔCRE). The method does not account for possible direct effects of aerosols above clouds, but the direct effects as a whole are small.

Table 2. Table of Changes to Quantities Between Indicated Case and Base Case[a]

Run (size, nm)	ΔTOA (mWm^{-2})	ΔSWCRE (mWm^{-2})	ΔLWCRE (mWm^{-2})	ΔCRE (mWm^{-2})	ΔR_{SWclr} (mWm^{-2})	ΔCLD (%)	ΔLWP (g m^{-2})	ΔIWP (g m^{-2})
BC-0.1%(38)	8	7	1	8	0	–0.002%	0.00	0.00
SO4 (8)	**–164**	**–255**	**95**	**–160**	**–11**	0.073%	0.39	0.05
SO4 (14)	**–46**	**–59**	**15**	**–44**	–3	0.021%	0.11	0.01
SO4 (21)	**–23**	**–25**	3	**–22**	–1	0.011%	0.04	0.00
H2O	**16**	**–20**	**40**	**20**	0	–0.015%	–0.01	0.03
ALL	**21±11**	**–81±12**	**63±5**	**–18±11**	2±1	0.01%	0.10±0.01	0.04±0.0
BC: 2% Eff								
BC-2% (11)	**–96**	**–321**	**224**	**–97**	8	0.068%	0.29	0.21
BC-2% (38)	–2	**–17**	**13**	–4	2	0.004%	0.01	0.01
BC-2% (79)	–1	–6	2	–4	3	0.001%	0.00	0.00
BC: Direct								
BC (11)	**–102**	**–147**	**47**	**–100**	–5	0.042%	0.25	0.03
BC (38)	–3	–6	2	–4	1	0.002%	0.01	0.00
BC (79)	–8	**–11**	2	–9	1	0.002%	0.01	0.00

[a]Included are net TOA radiation (ΔR), cloud radiative effect for SW (ΔCRE_{SW}), LW (ΔCRE_{LW}) and net (ΔCRE), clear sky TOA SW (ΔR_{SWclr}), cloud fraction (ΔCLD), liquid water path (ΔLWP) and ice water path (ΔIWP).
Significant global values (> $\pm$10 mWm^{-2} in bold.

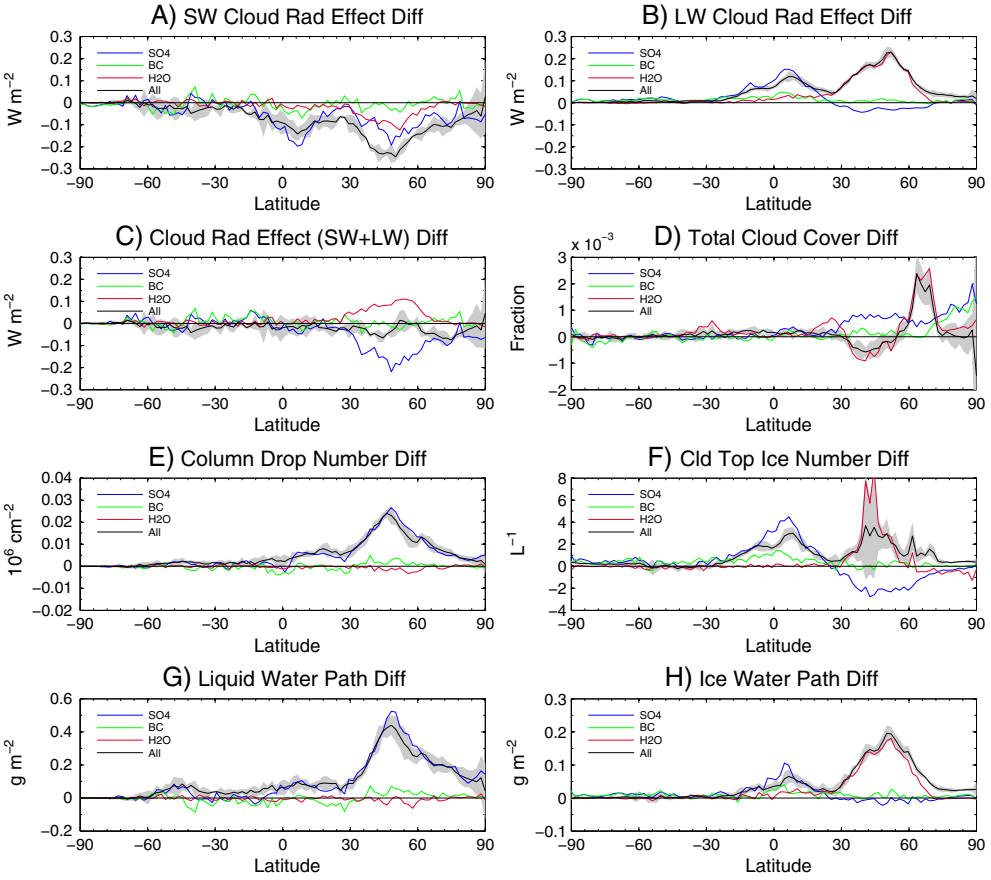

Figure 1. Zonal mean differences from Base case for SO_4 (blue), BC (green), H_2O (contrails: red) and "ALL" forcings (black). Differences for (a) shortwave, (b) longwave and (c) net cloud radiative effect as well as (d) total cloud cover, (e) column drop number, (f) cloud top ice number (g) liquid water path (LWP), and (h) ice water path (IWP). Gray shading for ALL forcings represents 2σ based on anomaly differences of four ensemble simulations with different meteorology.

3. Results

[12] Basic results of the simulations are shown as zonal mean differences from the base case in Figure 1, and as global differences in Table 2.

[13] The BC (black carbon) case with 0.1% efficiency (closest match to observations) has no significant variation from the base simulation in Figure 1 (green). Global results for BC (Table 2) are below the 10 mWm^{-2} significance threshold for direct ΔR_{SWclr}, indirect (ΔCRE), or combined (ΔR) effects. There is no significant change in ice number concentrations at cloud top due to BC (Figure 1f). Changes in number concentration due to BC are < 5%, consistent with *Hendricks et al.* [2011]. Assuming a higher efficiency (2%) for ice nucleation results in slightly more cooling, but the results are still not significant (Table 2). Statistically significant effects are found only for small (11 nm) BC sizes (Table 2) as a result of changes to liquid clouds: increased cloud fraction and liquid water path (Table 2). Previous results [*Penner et al.*, 2009] of large effects with aircraft soot are more consistent with the BC-2% 11 nm size simulation because earlier results assume the following: (1) high efficiencies for ice nucleating properties of soot (higher than observations [*DeMott et al.*, 2009]) and (2) high ice crystal and BC aerosol numbers (i.e., smaller sizes).

[14] Aviation SO_4 has significant impacts in Table 2. There is a global cooling of –46 mWm^{-2}, induced by cloud forcing of –44 mWm^{-2}. Direct effects (–3 mWm^{-2} from ΔR_{SWclr}) are consistent with *Lee et al.* [2009]. Indirect (cloud) forcing is concentrated in the northern hemisphere midlatitudes (Figure 1c: blue), and is almost exclusively a SW effect (Figure 1a: blue). In the tropics, the SW offsets the LW (Figure 1b) cloud radiative effect. Aviation SO_4 causes very small increases in total cloud cover (Figure 1d) due to changes in high clouds. Significant radiative effects for SO4 and other cases occur through changes to liquid clouds by increasing column drop number (Figure 1e) and liquid water path (Figure 1g). Zonal mean SO_4 mass increases by ~1% between 700 and 40 hPa in midlatitudes, increasing liquid drop number and liquid water path at 700 hPa and ice water path at 400 hPa. Liquid and ice particle sizes do not change significantly. The SO_4 is settling, advected, or scavenged and re-evaporated into regions where it affects liquid clouds. There are also changes to ice number (Figure 1f) and ice water path (Figure 1h), which perturbs tropical radiative fluxes (Figures 1a and 1b), but these largely cancel in the net ((Figure 1c). Aviation SO_4 is altering liquid clouds, by adding sulfate to serve as cloud condensation nuclei at altitudes below emission.

[15] We have performed sensitivity tests varying the assumed size (the inverse of the number concentration of particles emitted) based on the recommended ranges in *Barrett et al.* [2010]. For small (8 nm) SO_4 particles from

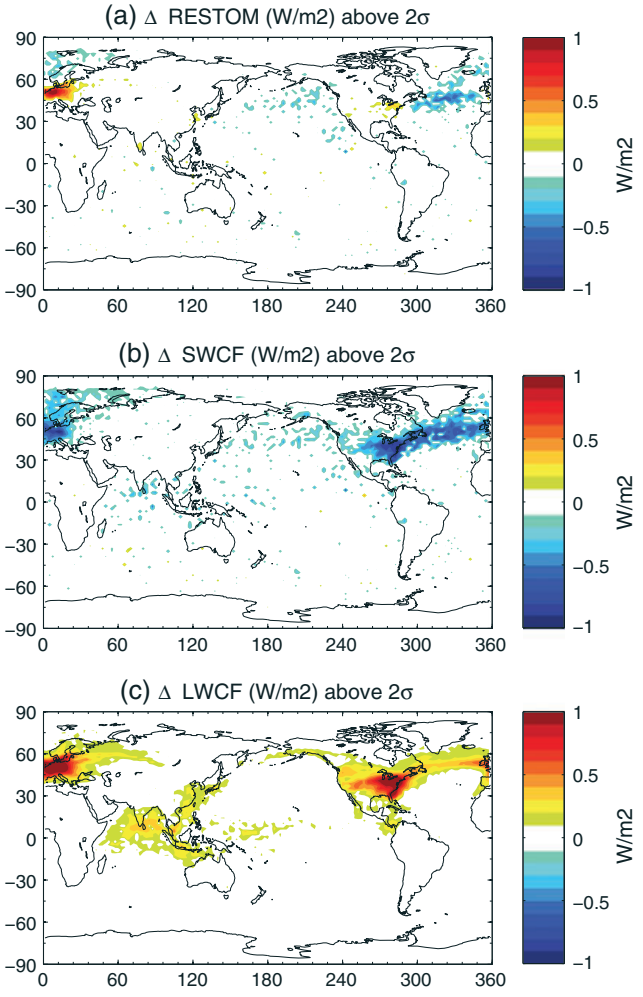

Figure 2. Differences between ALL and base case for (a) net, (b) shortwave and (c) longwave cloud radiative effect. Locations with anomalies > 2σ based on variability across four ensembles are colored.

aircraft, a large (–164 mWm^{-2}) cooling effect through interactions with liquid clouds occurs (Table 2). The effect is smaller if the sizes are larger (–23 mWm^{-2}) because the primary interaction is through the number of sulfate aerosols that become cloud condensation nuclei. With fixed mass, the number change is larger for smaller particles. The effect of added SO_4 is sensitive to the balance between homogenous and heterogenous ice nucleation [*Penner et al.*, 2009].

[16] Next we examine a simulation with contrails only. The H_2O simulation with just contrails and no aviation aerosols indicates a total warming due to contrails of 16 mWm^{-2} with monthly resolved emissions (Table 2). This occurs through enhancements to LW cloud forcing (Figure 1b) in midlatitudes, mostly concentrated over continental flight corridors over eastern North America and Central Europe. There are significant increases in cloud cover at higher latitudes (60°–70°N; Figure 1d) that do not contribute to the change in cloud radiative effect because the clouds there are optically thin. There are small increases in ice number (Figure 1f), but significant increases in ice water path (Figure 1h). No changes to the liquid phase are seen due to contrail emissions (Figures 1e and 1g).

[17] Combining the aerosol (BC and SO_4) and contrail effects (ALL) yields a consistent picture with the individual contributions. Four simulations with different meteorology from CAM are averaged and a mean (and 2σ uncertainty) reported. Global impacts are broadly linear in Table 2: a cooling from SO_4 (–46 mWm^{-2}) via liquid clouds offsets a warming from contrails (16 mWm^{-2}) and a BC warming (8 mWm^{-2}), with the cooling dominating when all effects are included (–21 mWm^{-2}).

[18] In the zonal mean (Figure 1, from the ensemble member with the same meteorology as the other experiments), this superposition is clear. The liquid effects on drop number (Figure 1e) and water path (Figure 1g) are almost entirely due to sulfate, while the ice effects (Figures 1f and 1h) are due to contrails (H_2O). The resulting cloud effect changes for the SW (Figure 1a) and LW (Figure 1b) nearly cancel (Figure 1c), but the net result is a small cooling, as would be expected from the larger SO_4 than contrail (H_2O) effect.

[19] Figure 2 illustrates the geographic structure of the total radiative effect at the Top of the Atmosphere (ΔTOA), which is approximately equal to ΔCRE, because global direct effects (ΔR_{SWclr}) are small (< 1 mWm^{-2}). In Figure 2, only locations with anomalies > 2σ based on variability across four ensembles are colored. The effects are concentrated over the highest air traffic corridors over N. America and Europe. SW effects (Figure 2b) are due to both aviation H_2O (contrails) and SO_4. Local perturbations to the local SO_4 budget from aircraft reach 1%–2%. The combined LW impact (Figure 2c) of contrails (H_2O) and aerosols looks very similar to the effect of contrails alone.

[20] The resulting net effect (Figure 2a) indicates that the SW cooling (due to SO_4 effects on liquid clouds) cancels out the LW warming from contrails over N. America, and partially over Europe. The net global effect is cooling. Figure 2 clearly shows the statistically significant signal is coherent, with regional net cooling effects of 200–300 mWm^{-2} over the N. Atlantic, and warming over Europe of 500 mWm^{-2}.

4. Conclusions

[21] Aviation BC exerts significant radiative forcing impacts only for the smallest assumed sizes (high number concentrations), and only due to interactions with liquid clouds. Direct effects of aviation aerosols are not significantly different from zero, and indirect effects make up most of the total response. Effects due to ice nucleation are not significant if a typical efficiency of 0.1% is assumed. This differs from previous work by *Penner et al.* [2009] due to very different nucleation efficiencies and BC number concentrations. An extreme (high efficiency and high BC number case) has comparable radiative forcing. SO_4 emissions are found to alter liquid clouds: contributing to cloud SW brightening through enhanced liquid water path and drop number concentrations from 700–400 hPa in major flight corridors below the main aviation emission level at ∼200 hPa, particularly in the N. Atlantic.

[22] Direct and (mostly) indirect effects on liquid clouds from SO_4 of –46 mWm^{-2} are larger than the warming effect due to contrail cirrus and aviation induced cloudiness (16 mWm^{-2}). The –46 mWm^{-2} represents about 3% of the –1600 mWm^{-2} total anthropogenic SW liquid cloud indirect effects in CAM5 [*Gettelman et al.*, 2012], significantly larger than

the 0.3% increase in SO_4 concentration in the N. Hemisphere due to aircraft. Increased efficacy of aviation SO_4 may result from concentration of the signal in the N. Atlantic region, which does not see much change in cloud radiative effect due to anthropogenic emissions from 1850 to 2000 [*Gettelman et al.*, 2012; Figure 6a].

[23] Including all effects in the model produces a cooling of -21 ± 11 mWm^{-2}. Aviation aerosol impacts are larger than warming due to contrails, and comparable to the aviation CO_2 radiative forcing of 30 mWm^{-2} [*Lee et al.*, 2009]. While the timescale of CO_2 radiative forcing is long (100 years or more), the timescale of contrails and contrail cirrus (hours to days or weeks) is more comparable to that for aerosol effects (about a week). These short-term effects (and their net cooling) should perhaps be considered together in making policy decisions on aviation emissions.

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