

## Effects of plume-scale versus grid-scale treatment of aircraft exhaust photochemistry

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[1] This is a study to examine the impact of modeling photochemistry from aircraft emissions in an expanding plume versus at the grid scale in an atmospheric model. Differences in model treatments for a single flight occurred at all altitudes during takeoff, cruise, and landing. After 10 h, the plume treatment decreased grid-scale ozone production by 33%, methane destruction by 30%, and carbon monoxide destruction by 32% at cruise altitude compared with the grid-scale treatment. The plume treatment changed the odd nitrogen partitioning by ~10%. For multiple overlapping flights at cruise altitude, final ozone, methane, and carbon monoxide perturbations decreased by 77, 68, and 74%, respectively, compared with the grid-scale treatment. Enhanced mixing with ambient air reduced the plume-scale and grid-scale differences. The persistent differences in photochemical activity indicate that individual plume treatment should be incorporated into 3-D modeling studies.

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### 1. Introduction

[2] This study compares the effect on tropospheric chemical composition of treating aircraft exhaust chemistry in an expanding and shearing plume versus at the grid scale in an atmospheric model. This issue is important since by far most 3-D global and regional atmospheric models to date have simulated the chemistry of aircraft-emitted chemicals assuming instantaneous mixing of the emissions to the grid scale. Such treatment, however, places emissions into grid cells that can be several orders of magnitude larger in volume than an early aircraft plume, potentially diluting the emissions and changing the associated chemistry. In particular, nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) from aircraft emissions affect ozone formation and destruction. Such ozone changes depend primarily on ambient and emitted  $\text{NO}_x$ , hydrocarbons, carbon monoxide (CO), sunlight, and temperature [Grooß *et al.*, 1998;

Moulik and Milford, 1999; Schumann, 1997]. Concentrations of chemicals also depend on the volume over which they are mixed. Several studies have suggested that treating chemistry at the grid scale, where the initial volume is large, might give results different than treating it at the plume scale [e.g., Meijer *et al.*, 1997; Petry *et al.*, 1998; Liang and Jacobson, 2000; Kraabøl and Stordal, 2000; Kraabøl *et al.*, 2000, 2002; Vohralik *et al.*, 2008; Cariolle *et al.*, 2009].

[3] Petry *et al.* [1998], Meijer *et al.* [1997], Kraabøl *et al.* [2000], and Cariolle *et al.* [2009] developed parameterizations to estimate effective emission indices, conversion factors, or reaction rates at cruise altitude that account for early plume chemistry [see Paoli *et al.*, 2011]. Kraabøl *et al.* [2002] used one such parameterization to solve chemistry among merged emissions from multiple aircraft in a single plume within each grid cell of a 3-D chemical transport model. They found that such treatment reduced ozone perturbations by 15–18% (1–1.5 ppb) at middle and high northern latitudes. Cariolle *et al.* [2009] also found a similar reduction in ozone. In contrast, Vohralik *et al.* [2008] implemented two parameterizations and found that the plume effects on ozone were between –5% and +5%, similar in magnitude to Meijer [2001]. Multiple overlapping plumes, as in a flight corridor, are likely to change the background atmosphere and thus chemical products that arise from subsequent flights. Global and regional models using parameterizations that neglect these overlapping plumes may not accurately capture the evolving chemistry in high-traffic corridors.

[4] This paper analyzes chemistry at the grid scale versus the plume scale by combining a subgrid plume computer representation with an accurate chemical solver. The purpose is to determine whether differences are sufficient to justify inclusion of the plume chemistry treatment in a 3-D global and regional climate model that separately tracks each individual aircraft flight worldwide [Jacobson *et al.*, 2011]. That model treats the subgrid evolution of aerosol and contrail particles within each aircraft plume, but gas chemistry of aircraft emissions at the grid scale. Chemistry within individual plumes in that model would be calculated using the same reaction set used at the grid scale. Exhaust from each aircraft would be emitted into the aircraft's own plume. Photochemistry would be solved in each plume until either a contrail that forms dissipates or the plume reaches the size of the grid cell, at which time, the emitted and chemically produced contents of the plume would be added to the grid scale. The work presented in this paper differs from previous plume studies in that it examines the effects of multiple, overlapping flights at cruise altitude by treating each flight as an individual plume. It also shows results for an entire flight, from takeoff to cruise to landing, demonstrating changes at different altitudes and flight modes, solves

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chemical equations using a Gear-type solver, which is considered to be highly accurate, provides results as a function of diffusion and shear parameters, and provides results for multiple chemicals.

## 2. Simulations

[5] A flight from Oakland to San Diego, CA (~800 km) on 25 August 2006 was modeled using chorded emission data from the Volpe National Transportation Systems Center [Wilkerson *et al.*, 2010]. Flight segments were grouped into takeoff, cruise, or landing groups, as described in the supporting information.

### 2.1. Single Flight

[6] Three initial simulations were performed to study the effects of plume-scale versus grid-scale treatment of photochemistry for a single flight. In case BG1, aircraft emissions were neglected and gas chemistry of background air alone was solved in 12 1 km thick layers (0–12 km) with 100 km × 100 km horizontal resolution. In case G1, aircraft emissions from a single flight were instantaneously mixed with background air to the grid scale in each of the same layers as in case BG1. In case P1, emissions were added to background air within a subgrid-scale plume that entrained outside air as it expanded and sheared over time. All cases were solved over 10 h. To isolate differences between modeling treatments, simulations did not include additional outside emission sources.

### 2.2. Multiple Flights

[7] A second set of three simulations was performed to investigate the effects of four flights passing through a flight corridor, one per hour for 4 h. The justification for multiple flights is based on a frequency analysis of plume intersections over California (Figure S7). Additionally, in situ observations by Schlager *et al.* [1997] measured emission signatures from 2 to 5 intersecting plumes aged between 0.25 and 3 h in the North Atlantic flight corridor. The 1 h delay was chosen because it allowed sufficient time to show the chemical products formed in the plume, but not so much time that production changes were dominated by diffusion. After 1 h, the plume volume was nearly 30 times that of the original plume, but 0.5% of the grid-scale volume. In the simulations, emissions were either neglected (case BG4) or added to the grid scale (case G4) or a plume (case P4) one flight per hour starting at time zero. In case G4, emissions were added to the grid scale and chemistry was solved for a total of 10 h. In case P4, the original plume was initialized with mixing ratios from ambient air plus emissions from the first flight, as in P1. The first flight entrained only ambient air as it expanded. After an hour of chemistry and plume expansion, emissions from the second flight were added as a new layer within the older plume. This process was repeated for the remaining two flights so that each new plume expanded into the exhaust of the flight preceding it. To conserve mass, each flight volume was mutually exclusive, such that the added concentration flux of an expanding layer was also subtracted from the layer into which it expanded. At each time step, concentrations in each layer were adjusted first due to chemistry and then due to volume-weighted fluxes between layers. At a given simulation time, concentrations were integrated across all nonoverlapping concentric plume rings to give an overall impact.

### 2.3. Plume Model Description

[8] At the start of each flight simulation, emissions were instantaneously added to a single homogeneous plume in each vertical layer. The plume at each layer had an elliptical cross section with semimajor axis  $a$ , semiminor axis  $b$ , and rotational angle  $\theta$ . These parameters were modeled to change with time due to diffusion and wind shear via a subgrid plume model [Naiman *et al.*, 2010]. Initial dimensions of  $a=100$  m,  $b=50$  m, and  $\theta=0^\circ$  were chosen to represent the plume size at the beginning of the dispersion regime [Naiman *et al.*, 2011]. Horizontal and vertical diffusion coefficients for all layers were assumed for simplicity to be  $D_h=20$  m<sup>2</sup>/s and  $D_v=0.158$  m<sup>2</sup>/s [Dürbeck and Gerz, 1996; Schumann *et al.*, 1995]. Wind shear values were 5.6, 4.8, and 3.6 E−3 s<sup>−1</sup> for 0–3 km, 3–7 km, and 7–15 km, respectively [Beres *et al.*, 2002]. Results for varied wind shear and diffusion are provided in the supporting information.

### 2.4. Gas Photochemistry

[9] Gas photochemistry was solved among 167 species in 305 reactions (260 kinetic, 45 photolysis) using SMVGear II, an unconditionally stable, positive definite, and mass-conserving Gear-type chemical ordinary differential equation solver [Jacobson, 1998]. Initial mixing ratios and atmospheric parameters are listed in Tables S1 and S2. For simplicity, flight emissions in all layers were added at 18:00 GMT (11 A.M. local), and chemistry was solved immediately.

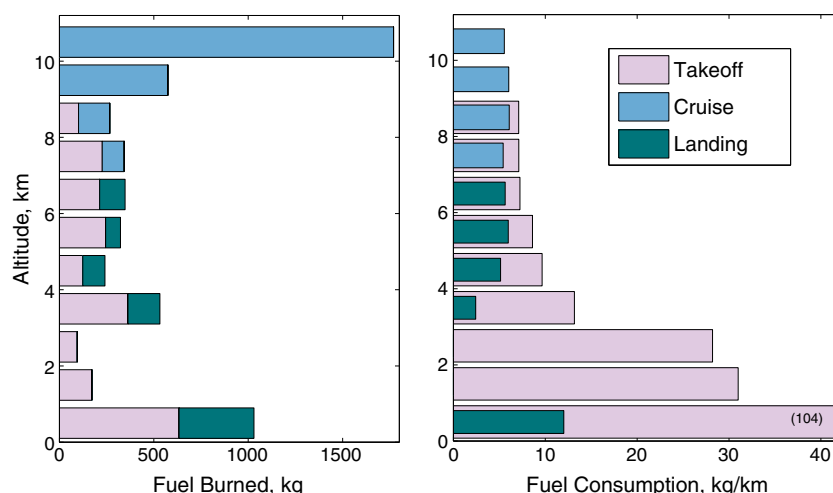
## 3. Results

### 3.1. Takeoff and Landing

[10] Figure 1 shows the amount of fuel burned for each of the takeoff, cruise, and landing scenarios. The cruise scenario burned the most fuel (2625 kg), followed by takeoff (2174 kg), then landing (896 kg). A significant portion of the takeoff and landing fuel burn occurred at ground level (<1 km), where 634 and 397 kg-fuel were consumed, respectively. Both takeoff and landing scenarios included times when the engine idled at low power settings on the ground (longer at takeoff than landing), making fuel consumption (kg-fuel/km-flown) highest for the two scenarios. At ground level, takeoff and landing emissions required 104 and 12 kg-fuel/km compared with 5–6 kg/km for the cruise segments above 7 km. Additionally, the higher thrust required for takeoff burned more fuel per kilometer than for landing at all altitudes.

[11] Hydrocarbons and carbon monoxide (CO) emissions, which result primarily from incomplete aircraft combustion, were highest at ground level where engine thrust settings were lower. Carbon monoxide contributed to daytime ozone formation through its reaction with the hydroxyl radical (OH) and formation of the hydroperoxyl radical (HO<sub>2</sub>). Although CO is an emitted species, this reaction contributed to a net CO loss ranging from −27 to −6 g/kg-fuel above ground level (>1 km), corresponding to a 19–71% decrease in grid-scale perturbations. These differences affect subsequent reactions with OH, which dictates the lifetime of many relevant photochemical species.

[12] In the plume, high nitric oxide (NO) titration initially reduced ozone, but subsequent nitrogen dioxide (NO<sub>2</sub>)



**Figure 1.** (left) Total fuel burned (kg) and (right) consumption rate (kg-fuel/km-flown) at each altitude and for takeoff, cruise, and landing scenarios.

formation and photolysis resulted in net ozone production at all altitudes (Figure 2). After 10 h, the plume treatment decreased ozone formation by 16–33% at all altitudes, relative to the grid-scale treatment. Generally, the plume treatment resulted in fewer  $\text{NO}_x$  conversions to reservoir species than the grid for a given time.

### 3.2. Cruise Altitude

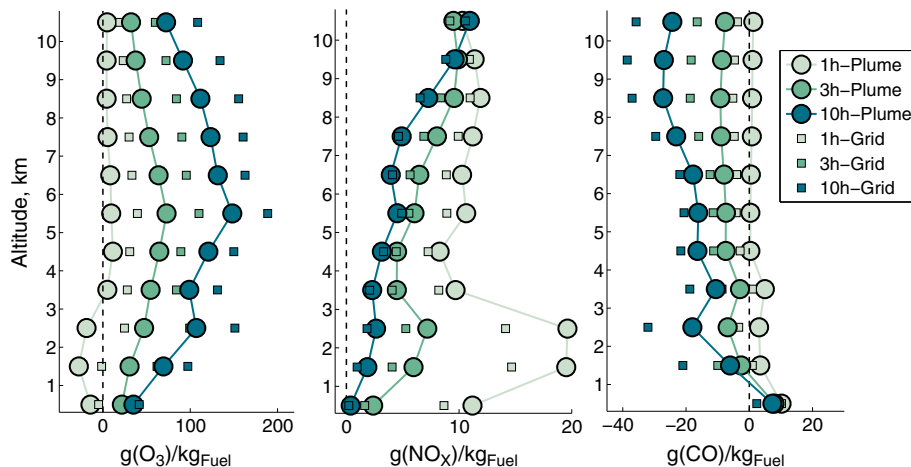
[13] The difference in early plume-scale versus grid-scale chemistry is particularly important at cruise altitude (10–11 km), where the flight burned the most fuel. The OH radical was produced primarily via the reaction  $\text{HO}_2 + \text{NO} \rightarrow \text{OH} + \text{NO}_2$ , which yielded more OH molecules at the grid scale than the plume scale. The higher absolute number of OH molecules resulted in more methane and carbon monoxide destruction by 30 and 32%, respectively, relative to the plume.

[14]  $\text{NO}$  titration at cruise altitude initially decreased plume ozone, followed by regeneration after 20 min as  $\text{NO}$  converted to  $\text{NO}_2$ , photolyzed, and produced ozone. The smaller plume volume also yielded higher  $\text{NO}_x$  mixing ratios, reducing the efficiency of  $\text{NO}_x$  to produce ozone [Liang and Jacobson, 2000; Jaeglé et al., 2001]. Grid-scale ozone,

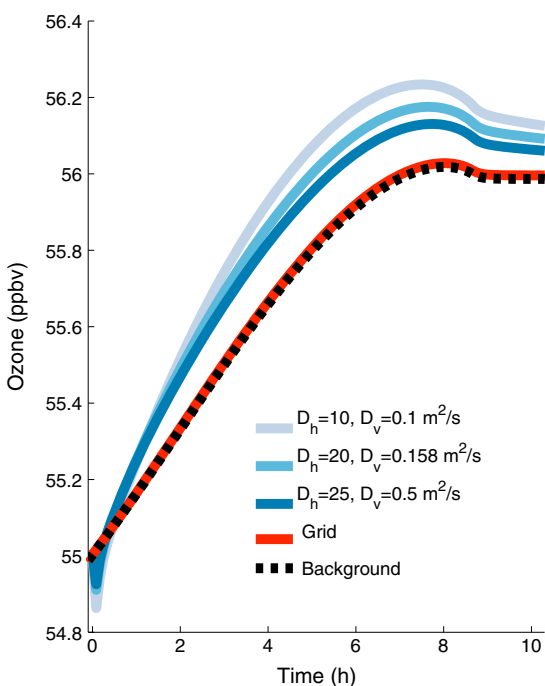
however, only increased, demonstrating a qualitative difference between the two treatments. After 10 h, the plume-scale treatment reduced ozone production from 109 g/kg-fuel at the grid scale to 72 g/kg, a 33% decrease. Under varying wind shear and diffusion scenarios, this decrease ranged from 25 to 55% (Figure S3). Increased turbulence diluted the plume with more ambient air, resulting in a larger plume with a lower ozone mixing ratio compared with the low turbulence scenarios (Figure 3). Overall ozone production, however, was highest with increased mixing since the dilution allowed for faster  $\text{NO}_x$  conversions. Using a layered plume would likely decrease plume mixing and further decrease ozone production [Meijer, 2001; Kraabøl et al., 2000].

### 3.3. Nitrogen at Cruise Altitude

[15] Nitrogen is emitted at cruise altitude as  $\text{NO}$ ,  $\text{NO}_2$ , and small amounts of nitrous acid ( $\text{HONO}$ ). The conversion of nitrogen into reservoir species is relevant to air quality, heterogeneous chemistry, particle formation, and future photochemistry. Since nitrogen is conserved in all simulations, it is useful to compare the  $\text{NO}_x$  conversion into reservoir species by tracking the emitted moles of nitrogen (= Mn).



**Figure 2.** Changes in  $\text{O}_3$ ,  $\text{NO}_x$ , and  $\text{CO}$  (g/kg-fuel) at each altitude when emissions are treated in an expanding plume (“Plume”, case P1) or at the grid scale (“Grid”, case G1) 1, 3, and 10 h after emission.



**Figure 3.** Ozone mixing ratios in the plume at cruise altitude (10–11 km) for low-, medium-, and high-turbulence conditions. Grid-scale and background mixing ratios are also shown.

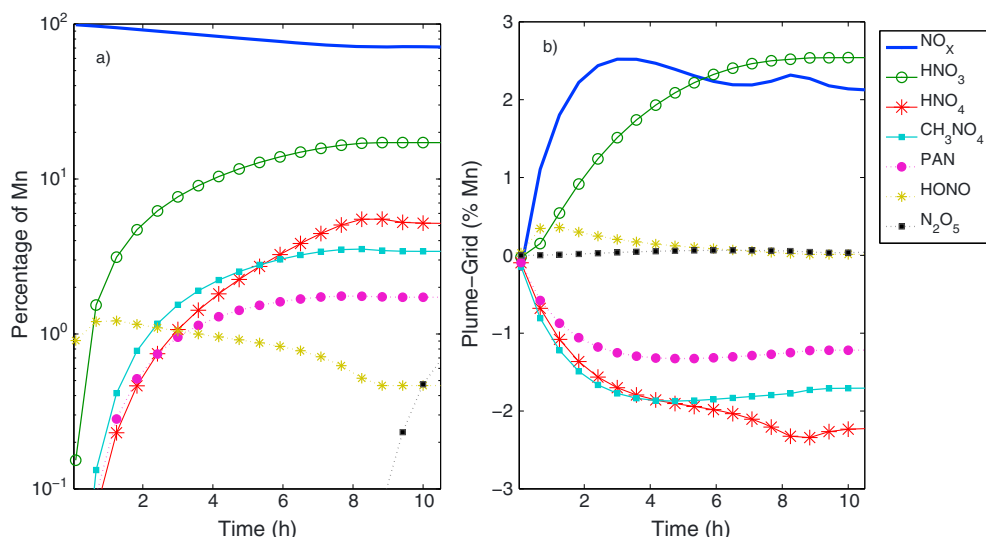
[16] Figure 4a shows that at cruise altitude, most of the plume nitrogen after 10 h (as a percentage of Mn) remained as  $\text{NO}_x$  (71%). The rest converted to nitric acid ( $\text{HNO}_3$ , 17%), peroxyacetyl nitrate ( $\text{PAN}$ , 1.7%), methylperoxy nitric acid ( $\text{CH}_3\text{NO}_4$ , 3.4%), or peroxyacetyl nitrate ( $\text{HNO}_4$ , 5.2%). This partitioning compares fairly well with the layered plume results of *Kraabøl et al.* [2000]. The grid-scale partitioning was similar, but with less remaining  $\text{NO}_x$  and  $\text{HNO}_3$  and more  $\text{HNO}_4$ ,  $\text{CH}_3\text{NO}_4$ , and  $\text{PAN}$  (Figure 4b). In terms of the total emitted moles of nitrogen, these differences are <3%, which appear small but can correspond to a

large change relative to an individual species. For instance,  $\text{PAN}$  mass production increased by 70% when treated at the grid scale, though it only represented 1.2% of the nitrogen molar partition. Similarly, more efficient reactions of  $\text{NO}_2$  with  $\text{OH}$  resulted in higher  $\text{HNO}_3$  production in the plume (18% increase, or 2.5% of Mn) compared with the grid-scale treatment. The differences in the plume-scale and grid-scale nitrogen did not diminish over the lifetime of the simulation (Figure 4b), demonstrating that the two treatments resulted in a persistent difference (~10% of Mn) in the nitrogen partition.

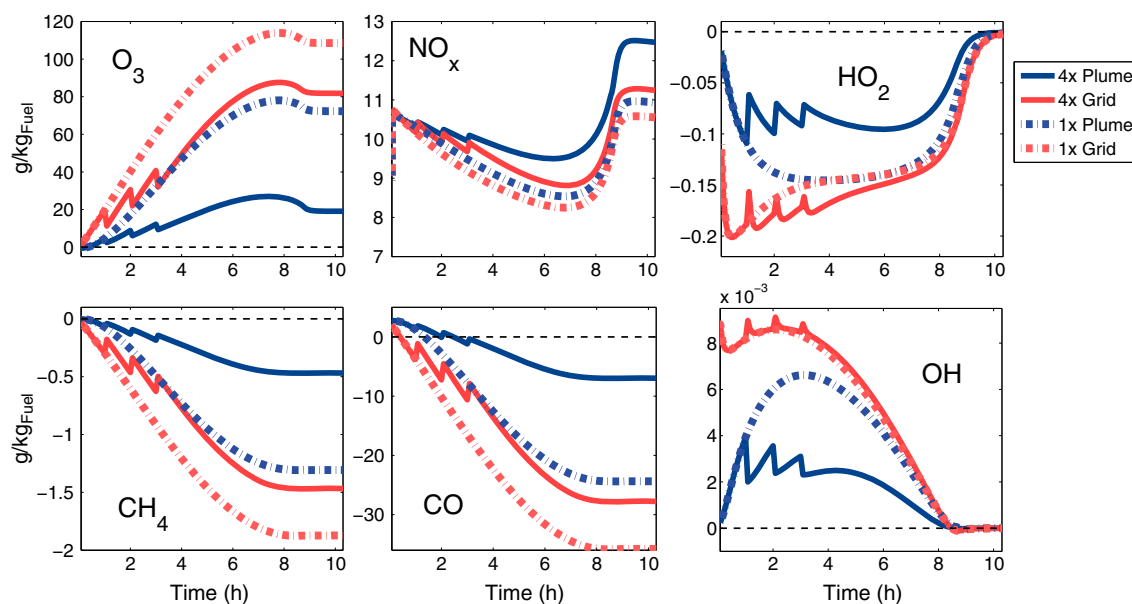
### 3.4. Multiple Flights

[17] Figure 5 shows the effects of multiple flights on chemistry in the plume versus grid scale at cruise altitude. Similar to the single-flight plume results, the additional flights at 1, 2, and 3 h also decreased ozone in the innermost layer, but decreases were not sufficient to overcome the ozone contribution from older flights. The additional fuel burned with each flight, however, decreased the total ozone to fuel ratio in both the plume and grid (Figure 5). Multiple flights decreased the ozone production ( $\text{g-O}_3/\text{kg-fuel}$ ) for both the plume-scale and grid-scale treatments. For the grid-scale treatment, the reduced production was primarily due to the reduced time for emissions to react before sunset, 8.7 h after the simulation start. For the plume treatment, the increasingly high  $\text{NO}_x$  mixing ratios within new plume layers further decreased ozone production efficiency with each flight. Thus, the multiple-flight plume treatment had the lowest ozone production among all simulations. These results agree with the work of *Kraabøl and Stordal* [2000], who modeled two flights by initializing a layered plume into the core of a plume aged 1 h. That study also found a reduction in  $\text{NO}_x$  conversions in the inner plume, as well as a reduction of ozone through the reaction with  $\text{NO}$ .

[18] After 10 h, the grid-scale treatment produced 82  $\text{g-O}_3/\text{kg-fuel}$ , and the plume treatment produced 19  $\text{g/kg}$ , a decrease of 77%. For a single flight, this was a 33% decrease, demonstrating the reduced ozone production efficiency with additional flights. Results were analogous for  $\text{CH}_4$  and  $\text{CO}$  destruction, which decreased by 68 and 74%, respectively, when multiple flights were treated in a plume.



**Figure 4.** Nitrogen in the plume at cruise altitude (10–11 km) (a) as a percentage of emitted moles of nitrogen (= Mn) and (b) as a difference (% of Mn) between plume-scale and grid-scale treatment.



**Figure 5.** Changes in select species (g/kg-fuel) at cruise altitude (10–11 km) when emissions are treated in an expanding plume (“Plume”) or at the grid scale (“Grid”) for a single flight (“1x”) or after four flights (“4x”).

#### 4. Discussion and Conclusions

[19] This study considered only gas-phase chemistry and neglected heterogeneous reactions that occur on contrail liquid and ice particles. Meilinger *et al.* [2005] found that in the presence of a persistent contrail, halogen activation and dehydrochlorination on ice particles resulted in a net loss of ozone from aircraft emissions. Furthermore, the relative plume-scale and grid-scale differences in chemical products are likely to change under different atmospheric conditions. In this study, background emission fluxes throughout the day were neglected, which could affect grid and plume differences, especially near the ground. However, this work showed that treatment differences are still likely to exist and that those differences will vary based on background conditions. The results of this paper should be seen as a demonstration of the potential effects of emissions dilution, rather than a realistic prediction of aircraft products.

[20] This study showed that treating aircraft emissions in an expanding plume generally slowed the conversion of  $\text{NO}_x$  to reservoir species, decreased ozone production, and decreased carbon monoxide destruction at all altitudes. At cruise altitude, the plume treatment also showed higher production of  $\text{HNO}_3$  but lower production of  $\text{HNO}_4$ ,  $\text{CH}_3\text{NO}_4$ , and PAN relative to the grid-scale treatment. Furthermore, for some species (e.g.,  $\text{O}_3$ ,  $\text{CH}_4$ , and  $\text{CO}$ ), the differences between the two treatments increased with time throughout the simulation. After 10 h, the plume treatment decreased ozone production (g- $\text{O}_3$ /kg-fuel) from a single flight by 33% and decreased methane and carbon monoxide destruction by 30 and 32%, respectively. Increased plume mixing with ambient air decreased these differences but did not eliminate them. For four overlapping flights, these differences in ozone, methane, and carbon monoxide were more pronounced, increasing to 77, 68, and 74%, respectively. These differences demonstrated an underlying change in subgrid-scale photochemical activity that is not accounted for in most global models. These changes are relevant to particle formation, heterogeneous chemistry, and future photochemistry,

which are all used to assess the impacts of aircraft emissions on atmospheric composition. Previous studies that have looked at the effects of aircraft on climate have added emissions to the grid scale or used parameterizations that do not directly track the evolving plume chemistry. These treatments do not account for multiple flight interactions or atmospheric conditions that vary within the lifetime of the plume. Results from this study indicate that including plume chemistry treatment in a model that tracks emissions from each flight separately, and can therefore account for varying atmospheric conditions, should improve the realism of 3-D climate model results.

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