The Impacts of Aviation Emissions on Current and Future Particulate Matter: The Effects of the Speciated Model Attainment Test on the Community Multiscale Air Quality Model Results

Matthew Woody

Research Advisor: Saravanan Arunachalam

School Affiliation: University of North Carolina at Chapel Hill

Degree status: Masters Candidate in Environmental Sciences and Engineering

Paper submitted to the Partnership for AiR Transportation Noise and Emissions Reduction (PARTNER): Joseph A. Hartman Student Paper Competition

February 7, 2010

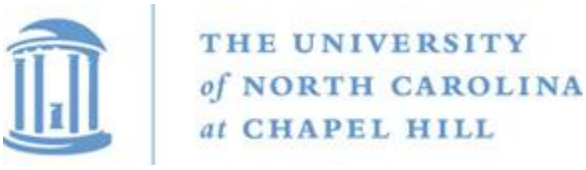

Table of Contents

Abstract

The impacts of aviation emissions on current and future year fine particulate matter were investigated using the Community Multiscale Air Quality (CMAQ) model and model results were post-processed using the Speciated Model Attainment Test (SMAT). CMAQ predicted aviation contributions to $PM_{2.5}$ in the U.S. were on average 0.037 $\mu g/m^3$ in 2005 and 0.0127 $\mu g/m^3$ in 2025 while SMAT predicted contributions of 0.0024 $\mu g/m^3$ in 2005 and 0.0096 in 2025 $\mu g/m^3$. SMAT typically predicted higher aviation contributions to sulfate aerosol while predicting lower aviation contributions to nitrate aerosol as compared to CMAQ results.

Introduction

Aviation is an integral part of daily global activities as an estimated 4.874 billion passengers traveled by aircraft in 2008 (Airport Council International, 2010). The Joint Planning and Development Office (JPDO), as part of the Next Generation Air Transportation System (NextGen), has suggested the number of passengers traveling within the U.S.'s aviation sector could double or even triple by 2025. With the increase in passenger traffic, this raises the important question as to what impact this will have on air quality as well as public health. Here we present an investigation of the impacts of aviation emissions on a current year (2005) and future year scenario (2025), focusing on $PM_{2.5}$ (fine particulate matter less than 2.5 micrometers in diameter), using the Community Multiscale Air Quality (CMAQ) model and post-processing the CMAQ results through EPA's Modeled Attainment Test Software (MATS) to perform the Speciated Model Attainment Test (SMAT). Because SMAT is routinely used in attainment demonstrations and health impact assessments in the U.S., we compare the air quality concentrations before and after applying SMAT to assess the significance of SMAT for aviation applications. The primary objective of this work is to quantify the influence of SMAT on the $PM_{2.5}$ concentrations attributed to aircraft emissions.

The state of the art, comprehensive, one atmosphere, regional air quality model, CMAQ, was used to quantify the impacts of aircraft emissions on air quality. In the case of $PM_{2.5}$, CMAQ provides the ability to model individual $PM_{2.5}$ speciated components: namely sulfate aerosol (SO_4), nitrate aerosol (NO_3), ammonium aerosol (NH_4), elemental carbon (EC), organic carbon (OC), and an "other" category (Crustal) which includes mostly unidentified crustal material.

SMAT is a model post-processor algorithm developed by the EPA that applies modeling data in a relativistic sense rather than an absolute sense to investigate air quality changes between two scenarios (U.S. EPA,

2007). To do this, SMAT uses the ratio between two modeling scenarios and applies these ratios to ambient monitoring data. SMAT has been used in a number of EPA policy relevant studies, such as regulatory impact analyses performed to support the Clear Skies, the Clean Air Interstate Rule (CAIR), and Low Sulfur Diesel Rule. Furthermore, the EPA requires states to apply SMAT in their State Implementation Plan (SIP) in conjunction with air quality modeling to demonstrate attainment of the National Ambient Air Quality Standards (NAAQS) for criteria air pollutants, including $PM_{2.5}$, as part of the Clean Air Act (U.S. EPA, 2007). SMAT was chosen because of its previous use in policy relevant work, because it is considered best practice by the EPA, its ability to produce speciated $PM_{2.5}$ fields, and its combination of ambient data and modeling results.

Methodology

CMAQ

A total of four annual air quality modeling simulations (Table 1) were performed in CMAQ over the continental U.S. at a 36-km grid resolution. 2005 meteorological inputs, which were applied in all four simulations, were generated from the PSU/NCAR mesoscale model (MM5). Model boundary and initial conditions for the current and future year scenarios were obtained from output generated by the GEOS-Chem global model (Bey et al., 2001). Aircraft emissions data generated from a research version of the FAA's Emissions and Dispersion Modeling System (EDMS), were then processed through the EDMS2Inv tool, and finally into the CMAQ emissions pre-processor, the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system (Baek et al., 2007). Non-aviation emissions for 2005 were obtained from the EPA's 2005 National Emissions Inventory (NEI). Future year non-aviation emissions were interpolated from EPA's 2020 and 2030 projection estimates and include projected growth and controls "on the books" for various sectors on the national and state level (U.S. EPA, 2008). Aircraft emissions for 99 major U.S. airports (Figure 1) were based on Terminal Area Forecasts (TAFs) for a single (typical) day as developed for JPDO in both the current and future years. National Airspace System (NAS) activity data in the current year and forecasts for the future year were obtained for this "seed" day using hourly emissions data, the values extrapolated to compute annual aircraft emissions inventories, and then the inventories were applied

Table 1. Summary of CMAQ Simulations

CMAQ Simulation	Description
base05	2005 case without aircraft emissions
airc05	base05 case plus 2005 aircraft emissions
base25	2025 case without aircraft emissions
airc25	base25 case plus 2025 aircraft emissions

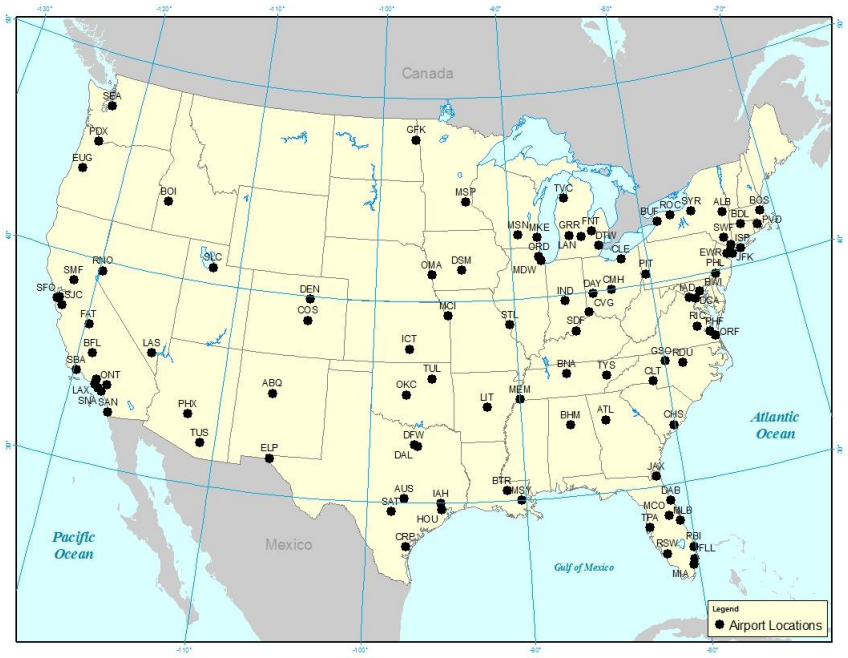

Figure 1. CMAQ modeling domain and location of 99 airports.

temporally and spatially using SMOKE. Future year aircraft emission estimates represent a "business as usual" scenario with no mitigation policies. These emissions are thus expected to provide a conservative estimate of potential growth in aviation emissions on future year air quality. Generally, background emissions decrease between 2005 and 2025, indicating an improvement of ambient conditions, while aviation emissions increase in the future (Figure 2).

SMAT

MATS is a modeling post-processor tool that uses speciated $PM_{2.5}$ CMAQ output to determine model predicted changes to ambient conditions by applying the SMAT process. SMAT results are available as point estimates and spatial estimates. Point estimates are calculated at CMAQ grid cells containing Federal Reference Method (FRM) monitoring sites while spatial estimates are calculated at each grid cell in the CMAQ domain.

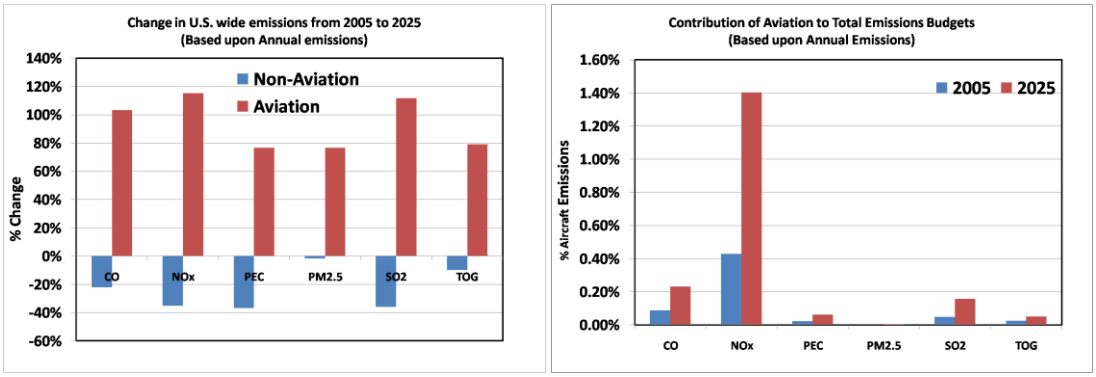

Figure 2. Change in Emissions from 2005 to 2025.

Table 2. Quarterly average $PM_{2.5}$ concentrations at LAX based on VNA interpolated FRM data from 2004 to 2006.

Quarter	1	2	3	4
$PM_{2.5}$ Mass ($\mu g/m^3$)	14.6178	12.0437	14.448	16.843

To illustrate the SMAT process, an example is given for the spatial estimate results for the CMAQ grid cell containing the Los Angeles International Airport (LAX). The first step in SMAT is to establish a baseline quarterly $PM_{2.5}$ mass (Table 2). This mass is obtained from the FRM air quality monitors and is the quarterly average typically calculated over a three year period. In CMAQ grid cells that do not contain a FRM monitor (as is the case with LAX), the FRM monitor data is spatially interpolated to the grid cell using Voronoi Neighbor Averaging (VNA), an inverse weighted nearest-neighbor technique (Abt Associates Inc., 2009). FRM monitors are used to determine attainment of the NAAQS and therefore used in the SMAT process to establish a baseline $PM_{2.5}$ mass. A primary difference in the FRM network and other $PM_{2.5}$ monitoring networks, such as the Speciated Trends Network (STN) and Interagency Monitoring of Protected Visual Environments (IMPROVE) network, is that only total $PM_{2.5}$ mass are available and not $PM_{2.5}$ speciation. To calculate speciated $PM_{2.5}$ concentrations at FRM monitors, speciated fractions are derived from STN and IMPROVE monitoring network data. These speciated fractions are calculated on a quarterly basis and are typically based on 3 years of monitoring data. Approximately 80% of FRM monitors are not co-located with STN or IMPROVE monitors and again interpolation is required (Figure 3) (U.S. EPA, 2006). For SMAT's spatial estimates, the VNA technique is used to interpolate speciated data to FRM sites.

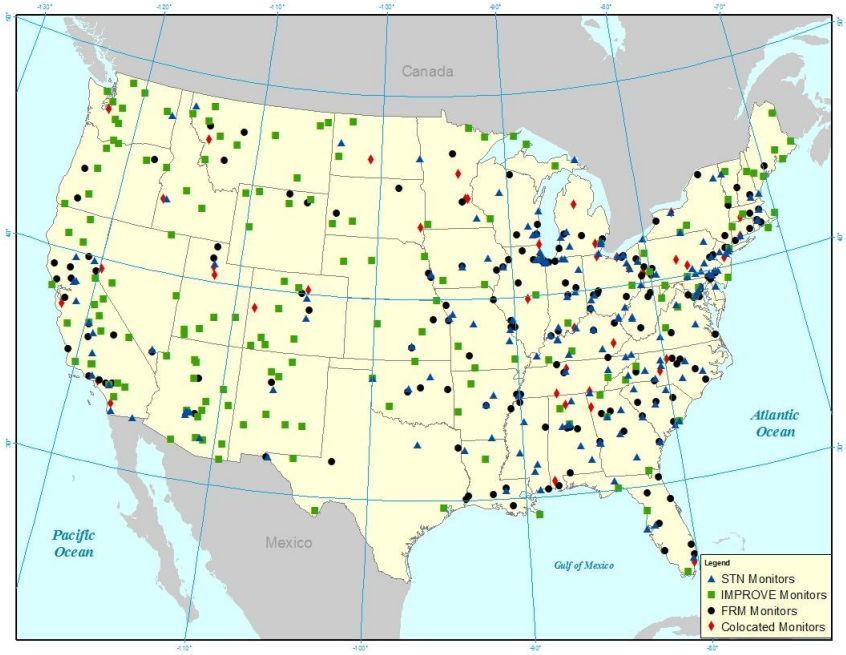

Figure 3. Location of Ambient Air Quality Monitors Used in SMAT.

There are, however, issues with using speciated data from monitoring networks due to limitations of the sampling methodologies. For example, sampling filters do not retain portions of volatile compounds, such as ammonium nitrate, and therefore lead to sampling artifacts (Frank, 2006). To alleviate this and other issues with sampling techniques, SMAT uses the sulfate, adjusted nitrate, derived water, inferred carbonaceous material balance (SANDWICH) technique developed by Neil Frank to calculate speciated $PM_{2.5}$ concentrations (Table 3). The speciation of SO_4, EC, and crustal material are relatively straightforward and the data from the speciated monitors can be directly applied. For NO_3, the reported speciated monitor data are adjusted to account for volatilization using a simple thermodynamic model (U.S. EPA, 2006). NH_4 is derived using a calculation based on the Degree of Neutralization (DON), or ratio of NH_4 neutralized by SO_4. DON is defined as

$$\mathrm{DON} = \mathrm{NH}_{4,\mathrm{SO}_4} / \mathrm{SO}_4 \qquad (1)$$

where NH_{4,SO_4} is NH_4 associated with SO_4 and SO_4 is measured SO_4. The DON calculation is required because NH_4 and SO_4 can combine to form ammonium sulfate ($(NH_4)_2SO_4$), ammonium bisulfate (NH_4HSO_4), or letovicite ($(NH_4)_3H(SO_4)_2$), depending on ambient conditions. NH_4 and NO_3 combine to form only ammonium nitrate (NH_4NO_3) and because of the one to one molar ratio, NH_4 associated with NO_3 can be calculated on a mass basis as

$$\mathrm{NH}_{4,\mathrm{NO}_3} = 0.29 * \mathrm{NO}_{3,\mathrm{Retained}} \qquad (2)$$

where NH_{4,NO_3} is the mass of NH_4 associated with NO_3 and $NO_{3,Retained}$ is the adjusted NO_3 mass as calculated by the thermodynamic model. NH_4 associated with SO_4 can then be calculated by taking the difference of measured NH_4 and NH_4 associated with NO_3, or

Table 3. a) Spatially estimated speciated fractions at the LAX grid cell as calculated by the SANDWICH technique using speciated monitoring data from 2004-2006. b) Spatially estimated speciated concentrations of FRM $PM_{2.5}$ mass at the LAX grid cell calculated by multiplying FRM $PM_{2.5}$ mass by speciated fractions.

a)

Quarter	Crustal (µg/m^3)	EC (µg/m^3)	NH_4 (µg/m^3)	OC (µg/m^3)	SO_4 (µg/m^3)	NO_3 (µg/m^3)	PBW (µg/m^3)
1	0.056	0.09	0.118	0.294	0.124	0.253	0.058
2	0.069	0.065	0.134	0.243	0.301	0.083	0.1
3	0.054	0.064	0.131	0.254	0.339	0.029	0.122
4	0.06	0.104	0.089	0.431	0.122	0.15	0.042

b)

Quarter	Crustal (µg/m^3)	EC (µg/m^3)	NH_4 (µg/m^3)	OC (µg/m^3)	SO_4 (µg/m^3)	NO_3 (µg/m^3)	PBW (µg/m^3)	$PM_{2.5}$ (µg/m^3)
1	0.7906	1.2706	1.6659	4.1506	1.7506	3.5718	0.8188	14.6178
2	0.7965	0.7503	1.5469	2.8051	3.4747	0.9581	1.1544	12.0437
3	0.7532	0.8927	1.8272	3.5428	4.7284	0.4045	1.7017	14.448
4	0.9806	1.6997	1.4545	7.0438	1.9938	2.4515	0.6864	16.843

$$NH_{4,SO_4} = NH_{4,measured} - 0.29 * NO_{3,Retained} \quad (3)$$

Finally, NH_4 can be calculated using the equation

$$NH_4 = DON * SO_4 + 0.29 * NO_{3,Retained} \quad (4)$$

The second speciated interpolation in SMAT is to calculate particle bound water (PBW). Ammonium sulfate and ammonium nitrate are hygroscopic and a portion of their mass as measured by ambient monitors include PBW (Abt Associates, Inc., 2009). PBW is derived from SO_4, NO_3, and NH_4 concentrations using a polynomial regression equation fit to data generated by the Aerosol Inorganic Model (AIM) (Clegg et al., 1998) (Abt Associates Inc. 2009.). The AIM PBW calculations were performed at ambient conditions of 35% relative humidity and 22 degrees Celsius, the conditions at which typical filter equilibration occurs (U.S. EPA, 2006).

Finally, because of uncertainties in estimating carbonaceous mass from carbon measurements and differences in carbon measurement protocol between urban (STN) and rural (IMPROVE) monitoring locations, OC is estimated in the SANDWICH technique (Abt Associates, Inc., 2009). To estimate OC, a mass balance approach is used that subtracts all other estimated species from the total FRM $PM_{2.5}$ measured mass using the equation

$$OC = PM_{2.5} - (SO_4 + NO_{3,Retained} + NH_{4,Retained} + PBW + Crustal + EC + Blank\ Mass + Salt) \quad (5)$$

Because there is a possibility equation 5 could calculate too large or small (or even negative) of a value for OC, OC is limited by both floor and ceiling values. The default floor value is set to 1 times the measured organic mass, based on the assumption that a portion of organic mass is volatile/semi-volatile and not completely retained on the filter (Abt Associates, Inc., 2009). The default ceiling value is set to 80% of the total $PM_{2.5}$ mass (Abt Associates, Inc., 2009). In cases where either the floor or ceiling values are used, all other $PM_{2.5}$ species are adjusted up or down by equivalent percentages to maintain a mass balance.

Once quarterly speciated masses are estimated for the base year using the SANDWICH technique, sensitivities derived from the model are applied to determine forecasted concentrations. In this application, the sensitivity case refers to CMAQ cases that include aircraft emissions (airc05 and airc25) and the base case refers to the CMAQ cases without aircraft emissions (base05 and base25). Because the only difference between the sensitivity and base cases are the addition of aircraft emissions, the differences between the two cases can therefore be defined as the contribution from aircraft emissions, or sensitivity of the model to aircraft emissions. In SMAT, this sensitivity is expressed as a relative reduction factor (RRF) as

$$RRF = \frac{\text{Model}_{\text{sens}}}{\text{Model}_{\text{base}}} = \frac{x_s}{x_b} \quad \text{(RRF can be > 1)} \qquad (6)$$

where $\text{Model}_{\text{sens}}$ is the speciated concentration as predicted by the model in the sensitivity case and $\text{Model}_{\text{base}}$ is the speciated concentration as predicted by the model in the base case (Table 4). The CMAQ-based RRF is then applied to SMAT's estimated baseline Crustal, EC, OC, SO_4, and NO_3 masses by multiplying each by their corresponding RRF to estimate SMAT concentrations in the sensitivity case (Table 5). For NH_4, the default approach for calculating forecasted concentrations is by applying the same DON value used in the base mass calculation to the forecasted SO_4 and NO_3 masses. The forecasted PBW is calculated by using the polynomial regression mentioned previously. This procedure is applied on a quarterly basis with the average of the four values serving as the annual average. Thus, at the end of the SMAT process, we have a difference in monitored values from the base year to a forecasted year (or scenario), based upon modeled changes.

Table 4. a) Quarterly averaged base05 and airc05 model based concentrations at the grid cell containing LAX. b) RRFs at the grid cell containing LAX as calculated by taking the ratio of the modeled sens case (airc05) concentration to the modeled base case (base05) concentration.

a)

Quarter	Crustal (µg/m^3)	EC (µg/m^3)	OC (µg/m^3)	SO_4 (µg/m^3)	NO_3 (µg/m^3)
base05 concentrations (2005 case without aircraft emissions)					
1	3.2855	1.3971	2.7157	1.3949	3.5015
2	2.4177	0.7919	1.9297	1.5882	2.8364
3	2.9115	0.9295	2.2282	1.6191	2.5491
4	4.2934	1.4373	3.4483	1.2118	2.8029
airc05 concentrations (base05 case plus aircraft emissions)					
1	3.2854	1.4019	2.715	1.3972	3.5117
2	2.4177	0.7964	1.9289	1.5904	2.8496
3	2.9115	0.9356	2.2274	1.6218	2.564
4	4.2933	1.444	3.4474	1.2136	2.8097

b)

Quarter	Crustal	EC	OC	SO_4	NO_3
1	1.0	1.0035	0.9997	1.0017	1.0029
2	1.0	1.0057	0.9995	1.0014	1.0047
3	1.0	1.0066	0.9996	1.0017	1.0058
4	1.0	1.0047	0.9998	1.0014	1.0024

Table 5. Spatially estimated values at LAX in 2005 with aircraft emissions as calculated by multiplying the SMAT base values by their estimated RRF.

Quarter	Crustal (µg/m^3)	EC (µg/m^3)	NH_4 (µg/m^3)	OC (µg/m^3)	SO_4 (µg/m^3)	NO_3 (µg/m^3)	PBW (µg/m^3)	$PM_{2.5}$ (µg/m^3)
1	0.7906	1.275	1.6719	4.1496	1.7535	3.5822	0.8226	14.6301
2	0.7965	0.7546	1.5457	2.8038	3.4795	0.9626	1.1542	12.0662
3	0.7532	0.8985	1.8231	3.5415	4.7364	0.4069	1.7032	14.4465
4	0.9806	1.7077	1.4494	7.0422	1.9967	2.4574	0.6914	16.8581

PBW Adjustment

Due to the methods for calculating and reporting particle bound water by CMAQ and SMAT, a direct comparison of $PM_{2.5}$ becomes difficult. Typically, CMAQ $PM_{2.5}$ is reported as dry $PM_{2.5}$, which excludes PBW. SMAT on the other hand, reports $PM_{2.5}$ as wet $PM_{2.5}$ mass and includes PBW. To further complicate the comparison, CMAQ uses the ISORROPIA thermodynamic model (Nenes et al., 1998) to determine inorganic apportionment and particle bound water whereas SMAT uses the AIM inorganic model. Particle bound water in CMAQ is calculated at the local ambient conditions for each time step and location of the model whereas SMAT calculates PBW at 35% relative humidity and 22 degrees Celsius on a quarterly averaged basis. Thus, CMAQ predicted PBW estimates are typically much higher than those estimated by SMAT. To better compare CMAQ and SMAT estimated PBW, box model simulations were performed using ISOREV (courtesy, Uma Shankar, UNC-IE), where ISORROPIA was run in reverse mode using CMAQ predicted concentrations of ammonium, sulfate, and nitrate from these scenarios to estimate PBW concentrations at the same ambient conditions used in SMAT. We used this mass of PBW to apportion between the nitrate-bound and sulfate-bound CMAQ aerosol concentrations (from each model simulation) to compute wet $PM_{2.5}$ concentrations that would be comparable with SMAT results.

Results

Continental U.S.

Results presented here indicate the change in annual $PM_{2.5}$ concentrations due to aircraft emissions. CMAQ results are the estimated contributions to $PM_{2.5}$ due to aircraft emissions, and hence are differences of modeled values. SMAT results are potential changes in ambient monitored concentrations due to a modeled change from the contribution of aviation emissions.

For CMAQ results within the continental U.S., 2005 aircraft emissions (airc05 minus base05) increased $PM_{2.5}$ concentrations on average by 0.0037 ug/m^3 (0.05% increase of total $PM_{2.5}$) and 2025 aircraft emissions (airc25 minus base25) increased $PM_{2.5}$ concentrations by 0.0127 ug/m^3 (0.21% increase of total $PM_{2.5}$). Nitrate was the largest speciated contributor in both comparisons, contributing on average 0.0019 ug/m^3 (0.16% increase in NO_3) and 0.0074 ug/m^3 (0.85% increase in NO_3) respectively (Figure 4).

SMAT point estimate results (at FRM monitored locations alone) for $PM_{2.5}$ concentrations in the continental U.S. indicated aircraft contributed on average 0.0036 ug/m^3 in 2005 (0.03% increase in total $PM_{2.5}$) and 0.0157 ug/m^3 (0.13% increase in total $PM_{2.5}$) in 2025. Sulfate was the largest speciated component in 2005,

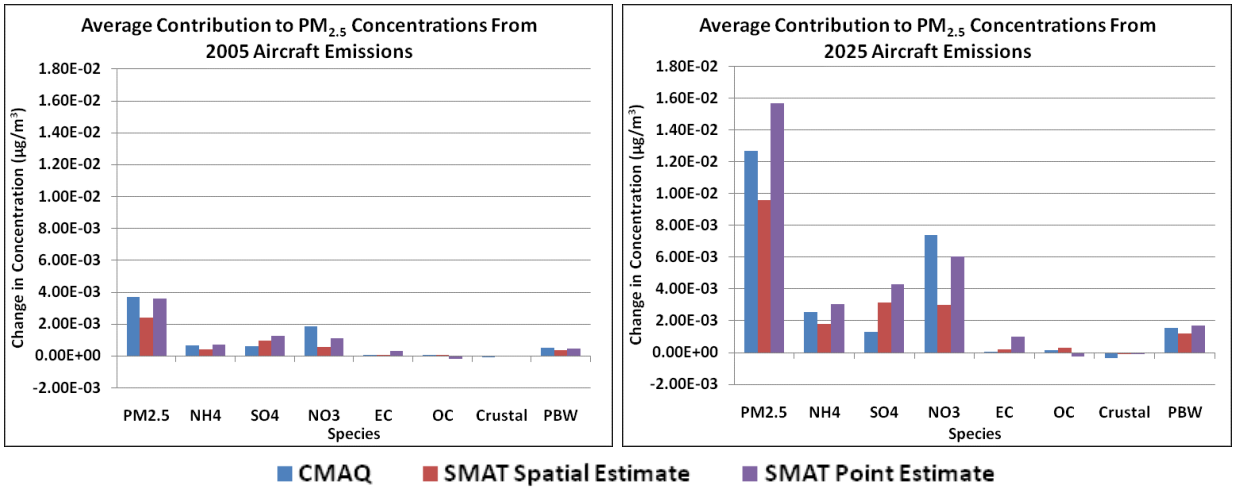

Figure 4. Average change in concentrations due to aircraft emissions in 2005 (left) and 2025 (right).

contributing an average of 0.0013 ug/m^3 (0.04% increase in SO_4). In 2025, nitrate was the largest speciated component, contributing an average of 0.0060 ug/m^3 (0.88% increase in NO_3) (Figure 4).

Spatial estimate SMAT results for $PM_{2.5}$ concentrations indicated an average increase of 0.0024 ug/m^3 (0.03% increase to total $PM_{2.5}$) due to aircraft emissions in 2005 and an average increase of 0.0096 ug/m^3 (0.11% increase to total $PM_{2.5}$) from 2025 aircraft emissions in the continental U.S. Sulfate was the largest speciated component in both years, contributing on average 0.0010 ug/m^3 (0.05% increase to SO_4) and 0.0032 ug/m^3 (0.15% increase to SO_4), respectively (Figure 4).

Figure 5 indicates regional differences in CMAQ and SMAT results for 2005 and 2025 by plotting the ratio of contributions from aircraft for spatial estimate SMAT results to CMAQ results. SMAT estimates of $PM_{2.5}$ contributions from aircraft are approximately one-fourth to three-fourths of those as predicted by CMAQ across much of the central and eastern portions of the U.S. Areas where SMAT results appear larger than CMAQ results occur primarily in the western U.S., notably in areas where SMAT predicts higher contributions of NO_3 from aircraft than those predicted by CMAQ. SMAT contributions from aircraft to NO_3 are sharply reduced as compared to CMAQ, particularly in the southeastern U.S., where SMAT predicted contributions of aircraft to NO_3 are approximately one-tenth of those as estimated by CMAQ.

Comparison of Hartsfield-Jackson Atlanta International Airport and Los Angeles International Airport

To better quantify the regional differences in CMAQ results after applying SMAT, presented here is a comparison of results from Atlanta Hartsfield International (ATL) (the busiest airport in the world based on 2008 passenger traffic) and LAX (the 6th busiest airport in the world based on 2008 passenger traffic) (Airports Council

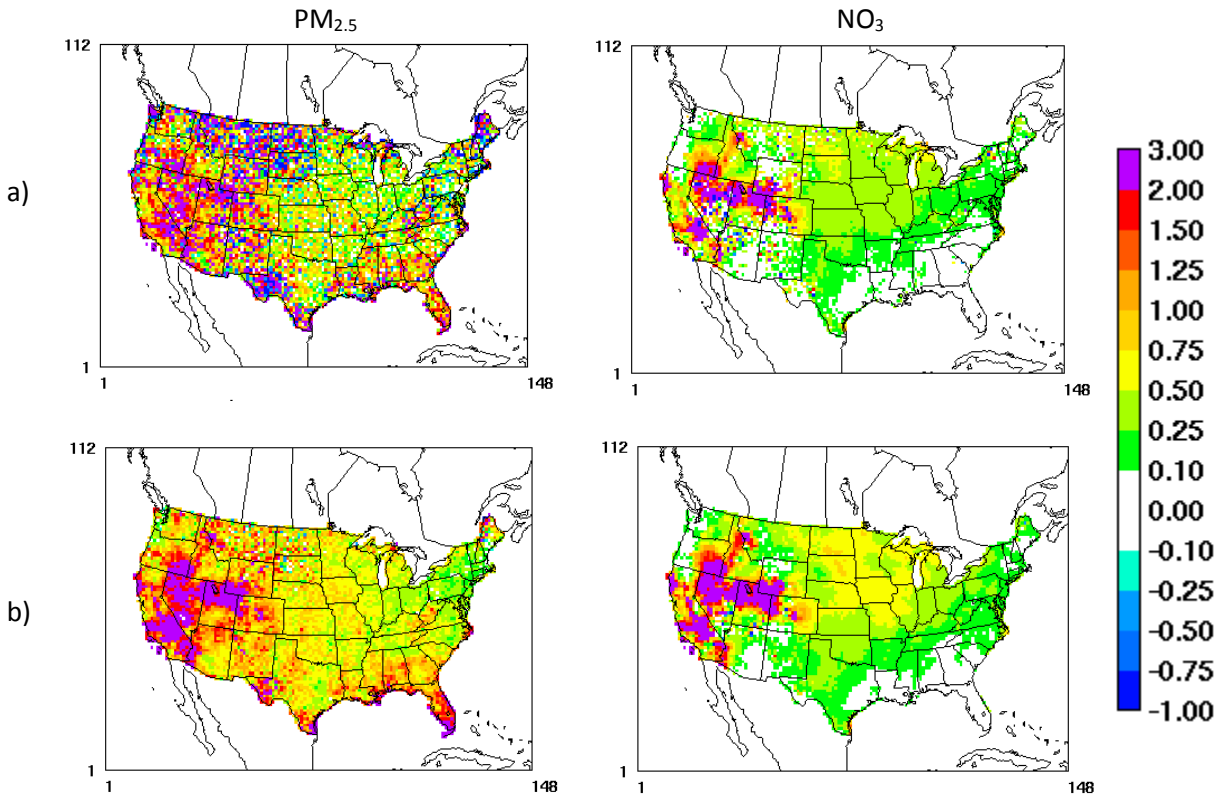

Figure 5. Ratio of changes due to aircraft as predicted by spatially estimated SMAT results to CMAQ results for $PM_{2.5}$ (left) and NO_3.(right), for a) airc05-base05, and b) airc25-base25

International, 2010). Additionally, these 2 airports are situated in locations that have distinct chemical regimes during the summer and winter seasons. In summer, the inorganic portion of $PM_{2.5}$ is dominated by sulfate in the eastern U.S. while sulfate and nitrate are approximately equivalent in the western U.S. (Bell et al., 2007). In winter, sulfate and nitrate are approximately equivalent in the eastern U.S. while nitrate dominates in the western U.S. (Bell et al., 2007).

At LAX, CMAQ $PM_{2.5}$ concentrations increased by 0.0245 ug/m^3 (0.18% increase to total $PM_{2.5}$) in 2005 and by 0.0560 ug/m^3 (0.46% increase to total $PM_{2.5}$) in 2025 due to aircraft emissions. NO_3 had the largest individual speciated contribution, with concentrations of 0.0113 ug/m^3 (0.39% increase to NO_3) and 0.0289 ug/m^3 (1.31% increase to NO_3), respectively. SMAT spatial estimates for $PM_{2.5}$ at LAX increased by 0.0162 ug/m^3 (0.11% increase in total $PM_{2.5}$) and 0.0524 ug/m^3 (0.36% increase in total $PM_{2.5}$), respectively, due to aircraft emissions. Again the largest contribution was attributed to NO_3, which increased by 0.0058 ug/m^3 (0.31% increase to NO_3) and 0.0190 ug/m^3 (1.03% increase to NO_3) respectively (Figure 6).

a)

b)

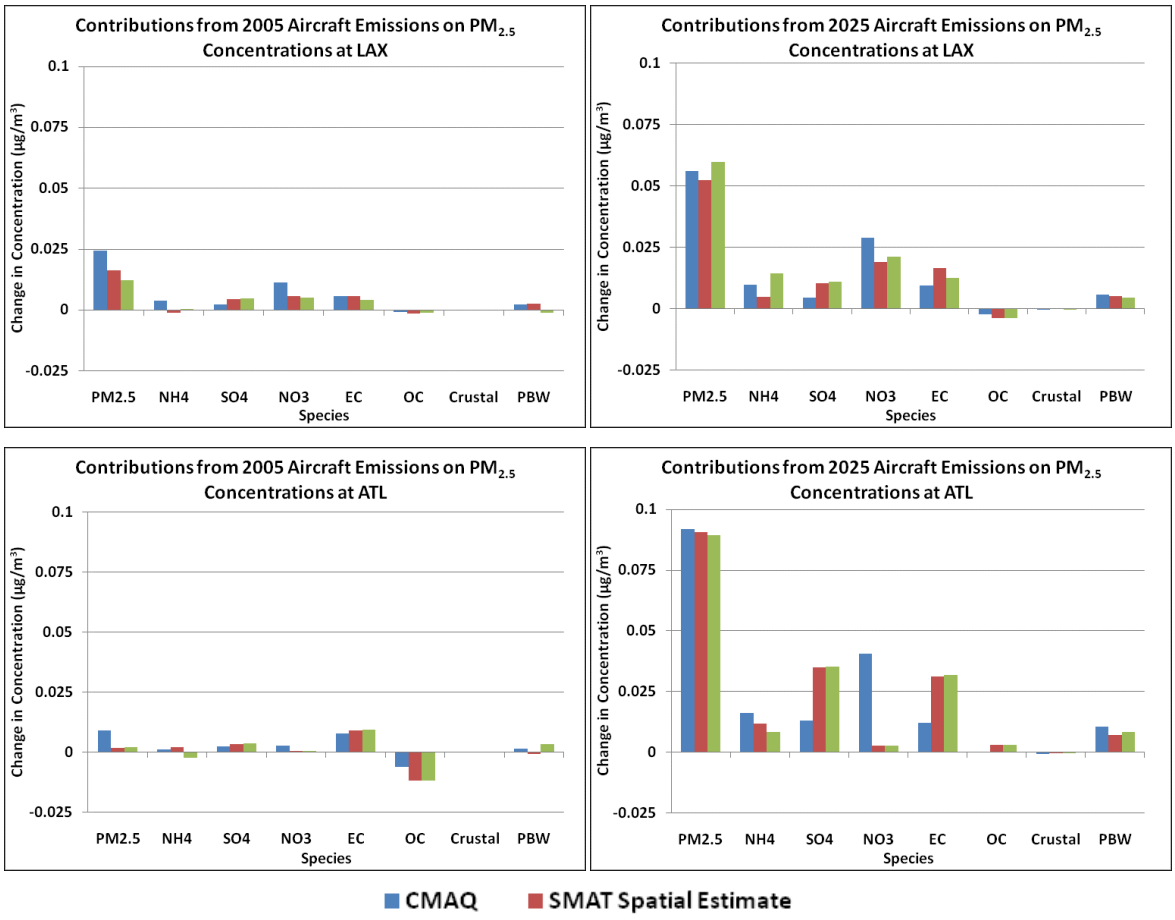

Figure 6. Change in $PM_{2.5}$ concentrations due to aircraft emissions in 2005 (left) and 2025 (right) at a) LAX, and b) ATL.

At ATL, CMAQ $PM_{2.5}$ concentrations increased by 0.0091 ug/m^3 (0.05% increase in total $PM_{2.5}$) due to 2005 aircraft emissions and by 0.0918 ug/m^3 (0.69% increase in total $PM_{2.5}$) due to 2025 aircraft emissions. EC contributed the most of any species in 2005, contributing 0.0021 ug/m^3 (0.89% increase to EC). In 2025, NO_3 had the largest contribution, contributing 0.0032 ug/m^3 (0.23% increase to NO_3) For SMAT spatial estimates, $PM_{2.5}$ increased by 0.0016 ug/m^3 (0.01% increase in total $PM_{2.5}$) and 0.0907 ug/m^3 (0.57% increase in total $PM_{2.5}$), respectively. EC had the largest contribution in 2005 with a concentration of 0.009 ug/m^3 (0.79% increase to EC). SO_4 had the highest contribution in 2025, with an increase of 0.0351 ug/m^3 (0.76% increase to SO_4) (Figure 6).

Discussion

On an annual average basis across the continental U.S., one of the primary differences in the contributions predicted by CMAQ and SMAT results is in the inorganic (NH_4, NO_3, SO_4) apportionment of $PM_{2.5}$ mass. CMAQ predicted that the largest speciated contribution from aircraft emissions was from NO_3 while SMAT results indicated that SO_4 had the largest speciated contributions to total $PM_{2.5}$ due to aircraft emissions. Although SMAT uses the

predicted changes to inorganic species from CMAQ (RRF values) to ultimately forecast the concentrations of the sensitivity cases, the differences in CMAQ and SMAT results can be attributed to differences in monitoring data and CMAQ predicted base concentrations. Figure 7 plots base case CMAQ concentrations against spatial estimate SMAT base concentrations (which are interpolated from ambient monitoring data) for NO_3, SO_4, and $PM_{2.5}$. $PM_{2.5}$ concentrations are roughly equivalent between the two in 2005 while CMAQ base concentrations appear lower than SMAT concentrations in 2025. Sulfate concentrations are typically higher in the SMAT base case than in the CMAQ base case while nitrate concentrations are typically lower for both 2005 and 2025. Also, SMAT base case estimates exhibit higher sulfate concentrations and lower nitrate concentrations overall. While ambient data indicate there are higher concentrations of sulfate than nitrate, uncertainties remain (and possible underpredictions) in nitrate measurements due to its volatility. This issue of NO_3 measurements has led to a lack of speciated $PM_{2.5}$ mass available outside of the U.S. and is one reason as to why SMAT is only applied in the U.S.

CMAQ has typically shown poor performance for predicting nitrate concentrations, overpredicting during winter months when conditions favor nitrate aerosol formation (Tesche et al., 2006). An analysis of the base05 case was performed using the Atmospheric Model Evaluation Tool (AMET), which compares modeled data against monitoring data, and indicated that CMAQ overpredicted NO_3 in winter months with both a high normalized mean bias (~75%) and high normalized mean error (~100%).

It is this combination of overall higher SO_4 to NO_3 concentrations in the SMAT base case as well as higher predicted SO_4 and lower predicted NO_3 concentrations as compared to the CMAQ base case which leads to the difference in organic apportionment. When an RRF is applied to a higher base value (SO_4), the contribution from

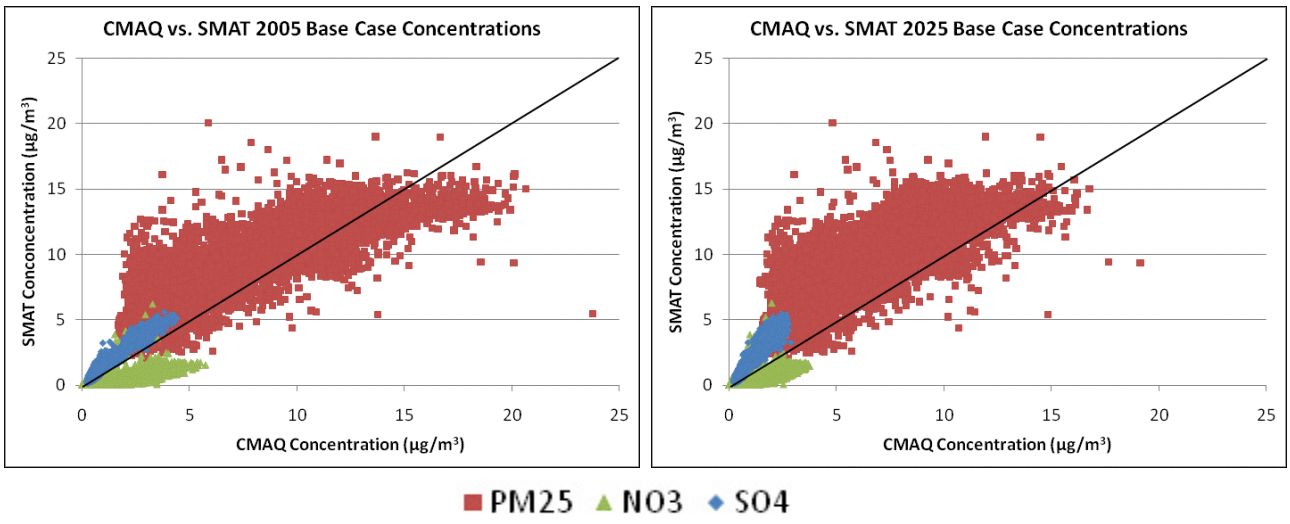

Figure 7. CMAQ vs. SMAT scatter plots of NO_3, SO_4, and $PM_{2.5}$ base concentrations in 2005 (left) and 2025 (right).

aircraft will be predicted to be higher than if it were applied to a lower base value (NO_3). Similarly, when an RRF value is applied to a higher base value in SMAT than that predicted by CMAQ, the overall contribution from aircraft as predicted by SMAT becomes greater than that predicted by CMAQ. The opposite is true for a lower value, the overall contribution from aircraft as predicted by SMAT becomes less than that predicted by CMAQ.

Comparing the overall changes in $PM_{2.5}$ mass, it is noteworthy that CMAQ as well as SMAT's point estimate exhibit larger total changes than those calculated by SMAT's the spatial estimate. The larger changes in the point estimate can be attributed to the fact that FRM monitors (the points) are typically located in urban areas and lead to an urban bias. Another contributing factor associated with the bias in point estimate is that the points are located near airports considered in the study.

The LAX and ATL comparison illustrates regional differences between CMAQ and SMAT results. The contributions of NO_3 from aircraft are essentially removed by performing SMAT at ATL, while at LAX they are simply reduced. This difference is attributed to ambient nitrate (as measured by the monitor) being significantly lower at ATL. Although similar RRF values for NO_3 are applied (1.0103 at LAX and 1.0194 at ATL for 2025), a smaller base value at ATL propagates to a smaller increase in the estimated contribution from aircraft. Also apparent in the comparison is the reduction of OC concentrations with the addition of aircraft emissions. Our previous investigations into this issue have indicated that aircraft emissions can cause reductions of OC in CMAQ, specifically the Secondary Organic Aerosol (SOA) component. Aircraft emissions at the airport react and remove free radicals that would otherwise participate in the creation of SOA (Woody and Arunachalam, 2009).

Results from this work were provided to the PARTNER's Project 11 team at the Harvard School of Public Health to quantify the health impacts of aircraft emissions as it relates to $PM_{2.5}$ contributions. Because differences in CMAQ and SMAT results cause changes in the relative importance of $PM_{2.5}$ speciated components, these speciated differences affect the health impact analysis. Since the magnitude of potential health impacts are used to identify and develop emissions control strategies, the use of CMAQ or SMAT results become significant as to which speciated component to prioritize to protect public health.

Conclusions

Aircraft emissions are found to increase $PM_{2.5}$ concentrations in 2005 and 2025, using both CMAQ output and those output processed by SMAT, with the largest contributions occurring in the future year. CMAQ predicted aviation contributions to $PM_{2.5}$ in the U.S. were on average 0.037 $\mu g/m^3$ in 2005 and 0.0127 $\mu g/m^3$ in 2025 while

SMAT spatially adjusted estimates predicted contributions of 0.0024 μg/m^3 in 2005 and 0.0096 in 2025 μg/m^3. The combination of higher amounts of aircraft emissions and lower background emissions in the future lead to the increased absolute contributions of $PM_{2.5}$ from aircraft.

The primary differences between predicted $PM_{2.5}$ contributions from aircraft emissions in CMAQ and SMAT are in the inorganic apportionment as CMAQ predicts NO_3 to be the largest speciated contributor (contributing on average 0.0019 ug/m^3 and 0.0074 ug/m^3) to $PM_{2.5}$ while SMAT predicts SO_4 as the largest contributor (contributing on average 0.0010 ug/m^3 and 0.0032 ug/m^3). SMAT also reduces the average $PM_{2.5}$ contribution from aircraft in its spatial estimate. SMAT point estimate results, with its clear urban bias, more closely resembles CMAQ predicted $PM_{2.5}$ aviation contributions. Based on these results, one might conclude that either CMAQ results overpredict or SMAT results underpredict the impact of aircraft emissions on changes to $PM_{2.5}$ concentrations. It is difficult to know which one of these is closer to the actual impacts due to aviation emissions. Furthermore, there are a number of obstacles making a direct comparison between the two difficult.

One such obstacle is that SMAT results include PBW in $PM_{2.5}$ concentrations and calculate PBW at standard conditions. However, we were able to address this key issue in this study, and facilitate a better comparison of CMAQ and SMAT results by including PBW in CMAQ $PM_{2.5}$ concentrations that were calculated at the same standard conditions as those SMAT.

One advantage to using SMAT is that it removes some of the uncertainties associated with modeling results, by focusing on using models in a relative sense. For example, the accuracy of emission inventories used in models is often questioned. By using SMAT, these uncertainties in the base emissions are reduced because the baseline $PM_{2.5}$ mass is based on ambient monitoring data. However, SMAT has its own set of uncertainties associated with it, such as the uncertainty involved in the volatilization of $PM_{2.5}$ mass and different sampling protocols between networks.

SMAT results are based on ambient measurements taken at monitoring locations across the U.S. and reflect ambient levels that populations are ultimately being exposed to. Therefore, SMAT results could be used to access potential health effects. However, as the results from this case study illustrate, the CMAQ contributions as compared to spatial estimated SMAT contributions indicate greater impacts on air quality. In fact, the CMAQ results are more in line with the urban biased SMAT point estimate results. Furthermore, the spatial analysis of CMAQ and SMAT results indicate that SMAT aviation contributions were smaller in the eastern U.S. (areas with

higher population densities) and higher in portions of the western U.S. (areas of lower population). Thus, SMAT results could cause health impacts to be biased low as compared to CMAQ results due to a higher proportion of the population being exposed to lower contributions of aircraft emissions to $PM_{2.5}$ concentrations.

There are limitations and assumptions both in CMAQ raw results and those post-processed using SMAT. For example, SMAT results are limited by ambient monitoring data available. It would be difficult to investigate scenarios where ambient conditions improve or worsen from currently sampled ambient conditions (e.g. future year conditions with increased regulations and better ambient conditions). CMAQ, on the other hand, represents the current scientific understanding of the environment, and while strides have been made, all atmospheric pathways of particulate matter formation are still not fully understood or incorporated in the model at this time.

Is post-processing CMAQ results using SMAT the best practice for determining contributions from aircraft emissions on $PM_{2.5}$ concentrations for performing health risk assessments? SMAT may be a valuable tool in determining current year contributions of aircraft emissions on air quality; however, in the case of future year air quality, ambient conditions may change significantly from current conditions, nullifying SMAT's advantage of results based on ambient conditions. Furthermore, SMAT is typically used to analyze emission reduction strategies on large scale emission sectors. Using it to analyze a relatively smaller emission sector, such as aircraft (compared to other anthropogenic sources), may stretch the limits of the tool. This is evident in the limited precision available in the algorithms used in SMAT where calculations are typically carried out to only 3 decimal places. For $PM_{2.5}$ concentrations, values are reported to 2 decimal places (which may not accurately capture the small incremental contribution from aircraft to $PM_{2.5}$) and require using the sum of speciated components instead of the reported $PM_{2.5}$ values to increase precision. For these reasons, while the SMAT process is a valuable tool that provides a better understanding of model results, we have identified several limitations in its current form for determining contributions of aircraft emissions to air quality, both present and future.

Future considerations of this work could include the effects of climate change on future year meteorological conditions and what impacts this has on the contribution of aircraft emissions to air quality. Another consideration would be to include aviation emissions of aircraft at cruising altitudes instead of only landing and take-off cycles as is currently modeled.

Acknowledgements

This work was funded by PARTNER, under Grant No. 07-C-NE-UNC, Amendment Nos. 001, 002 and 003. The Local Air Quality project is managed by Christopher Sequeira.

Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of PARTNER and its sponsors. PARTNER is funded by FAA, NASA, and Transport Canada.

I would also like to thank my advisors Sarav Arunachalam and Jason West for their help, insight, support, and time.

References

Abt Associates. Modeled Attainment Test Software User's Manual. Available at http://www.epa.gov/scram001/guidance/guide/MATS-2-2-1_manual.pdf, March, 2009.

Airports Council International. Available at: http://www.airports.org/. Accessed on January 21, 2010.

Baek, B.H., S. Arunachalam, A. Holland, Z. Adelman, A. Hanna, T. Thrasher, and P. Soucacos, Development of an Interface for the Emissions Dispersion and Modeling System (EDMS) with the SMOKE Modeling System, In Proceedings of the 16th Annual Emissions Inventory Conference - "Emissions Inventories: Integration, Analyses and Communication", Raleigh, NC, May 2007.

Bell, M.L., F. Dominici, K. Ebisu, S.L. Zeger, and J.M. Samet. Spatial and Temporal Variation in PM2.5 Chemical Composition in the United States for Health Effects Studies. Environmental Health Perspectives, Volume 115, Number 7, pp. 989-9985, 2007.

Bey, I., D.J. Jacob, R.M. Yanosca, J.A. Logan, B. Field, A.M. Fiore, Q. Li, H. Liu, L.J. Mickley, and M. Schultz. Global modeling of tropospheric chemistry with assimilated meteorology, Model description and evaluation. Journal of Geophysical Research, Volume 106, Number 23, pp. 23,073-23,095, 2001.

Clegg, S.L., P. Brimblecombe and A.S. Wexler. Aerosol Inorganics Model; A thermodynamic model of the system H+ - NH4+ - SO4- - NO3- - H2O at Tropospheric Temperatures. Journal of Physical Chemistry. 102A, pp

Frank, Neil. Retained Nitrates, Hydrated Sulfates, and Carbonaceous Mass in Federal Reference Method Fine Particulate Matter for Six Eastern U.S. Cities. Journal of the Air and Waste Management Association, Volume 56, pp 500-511, 2006

Nenes, A., S.N. Pandis, and C. Pilinis. A new thermodynamic equilibrium model for multiphase multicomponent inorganic aerosols. Aquatic Geochemistry, Volume 4, pp. 123-152, 1998.

Tesche, T.W., Ralph Morris, Gail Tonnesen, Dennis McNaly, James Boylan, and Patricia Brewer. CMAQ/CAMx annual 2002 performance evaluation over the eastern US. Atmospheric Environment, Volume 40, pp. 4906-4919, 2006.

U.S. EPA. Procedures for Estimating Future PM2.5 Values for the PM NAAQS Final Rule by Application of the Speciated Model Attainment Test (SMAT). September, 2006.

U.S. EPA. Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze. April, 2007.

U.S. EPA. Technical Support Document: Preparation of Emissions Inventories for the 2002-based Platform, Version 3, Criteria Air Pollutants. January, 2008.

Woody, Matthew and Saravanan Arunachalam. Secondary Organic Aerosol produced from aircraft emissions at the Atlanta Airport – an advanced diagnostic investigation using process analysis. In proceedings of the 8th Annual CMAS Conference, Chapel Hill, NC, October 2009.

Student Biography

Matthew is a M.S. candidate in the Department of Environmental Sciences and Engineering at the University of North Carolina at Chapel Hill and a UNC Institute for the Environment Fellow. As a second year student, he is currently working under Dr. Saravanan Arunachalam with the UNC Center for Environmental Modeling for Policy Development on the PARTNER Project 16 - Investigation of Aviation Emissions Air Quality Impacts. His chief research interests include atmospheric air pollution and air quality modeling.