Atmospheric Environment 79 (2013) 101–109

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Secondary organic aerosol produced from aircraft emissions at the Atlanta Airport: An advanced diagnostic investigation using process analysis

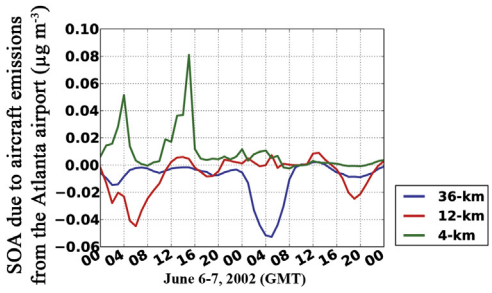
Matthew C. Woody [a,b], Saravanan Arunachalam [b,*]

[a] Department of Environmental Sciences and Engineering, Gillings School of Global Public Health, University of North Carolina at Chapel Hill, NC 27599, USA
[b] Institute for the Environment, University of North Carolina at Chapel Hill, 137 E. Franklin St., #648A, Chapel Hill, NC 27599-6116, USA

HIGHLIGHTS

- Impacts of aircraft LTO emissions on SOA modeled at the Atlanta Airport in CMAQ.
- Aircraft emissions reduced SOA at 36 and 12-km (by ~6%), but enhanced at 4-km (by ~12%).
- Process analysis used to determine processes responsible for changes in SOA.
- At coarser resolutions, chemistry of free radicals with aircraft NO_x emissions reduced SOA.
- At the finer resolution, aircraft emissions interacting with biogenic SOA precursors enhanced SOA.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 1 October 2012
Received in revised form
23 May 2013
Accepted 6 June 2013

Keywords:
Aircraft emissions
CMAQ
Secondary organic aerosols
Process analysis
Particulate matter
Grid resolution

ABSTRACT

Efforts using the Community Multiscale Air Quality (CMAQ) model to investigate the impacts of aircraft emissions from the Hartsfield-Jackson Atlanta International Airport have previously shown aircraft emissions increased total daily $PM_{2.5}$ concentrations by up to 9.4% (0.94 μg m^{-3}) with overall impacts varying by modeled grid resolution. However, those results also indicated that secondary organic aerosol (SOA) concentrations in the airport grid cell were reduced due to aircraft emissions at coarser grid resolutions (36-km and 12-km) but not at a finer resolution (4-km). To investigate this anomaly, this study instruments the CMAQ model with process analysis, an advanced diagnostic modeling tool, and focuses on changes to SOA concentrations due to aircraft emissions in the grid cells containing the Atlanta airport at grid resolutions of 36-km, 12-km, and 4-km. Model results indicated aircraft emissions reduced hourly anthropogenic and biogenic SOA concentrations at the 36-km and 12-km grid resolutions by up to 6.2% (0.052 μg m^{-3}) by removing nitrate, hydroxyl, and hydroperoxy radicals through chemistry. At the 4-km resolution, however, hourly modeled SOA concentrations increased (primarily due to changes in biogenic SOA) by up to 11.5% (0.081 μg m^{-3}) due to primary organic aerosol emissions from aircraft, with the additional organic mass shifting partitioning of SOA semi-volatile gas phase species into the particle phase.

* Corresponding author. Tel.: +1 919 966 2126; fax: +1 919 843 3113.
E-mail address: sarav@email.unc.edu (S. Arunachalam).

1352-2310/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.atmosenv.2013.06.007

1. Introduction

Aviation is an integral part of daily global activities, transporting approximately 725 million passengers and 67 million tons of cargo in the U.S. in 2011 over approximately 18 million flights (FAA, 2012a). The number of passengers flying is expected to grow by 3.1% per year through 2032 and eclipse 1 billion passengers in 2024 (FAA, 2012b). While important to transportation, aircraft contribute to both noise and air pollution. Aircraft are the third largest producer of greenhouse gas emissions (11.6% of the total) within the U.S. transportation sector behind light duty vehicles (58.7%) and freight trucks (19.2%) (Department of Transportation, 2010) and account for 3.5% of global anthropogenic radiative forcing (Lee et al., 2009). Furthermore, aviation activities have known emissions of CO, NO_x, volatile organic compounds (VOCs), SO_x, $PM_{2.5}$, and numerous hazardous air pollutants which adversely affect air quality (Brasseur et al., 1998; EPA, 1999; Wilkerson et al., 2010). In 2006, global commercial aircraft activities emitted approximately 2.7 Tg of NO_x (as NO_2-equivalent), 0.68 Tg of CO, 0.10 Tg of hydrocarbons (as CH_4-equivalent), 0.038 Tg of black carbon, 0.0041 Tg of primary sulfate aerosols, and 0.0028 Tg of primary organic aerosols (Wilkerson et al., 2010; Olsen et al., 2013). Ambient measurements have indicated that aircraft emit nanoparticles, with emissions (on the order of 10^{15}–10^{17} particles per kg of fuel burn) comparable on a per unit fuel burn basis to the number of particles generated from ship emissions, biomass burning and forest fires (Kumar et al., 2013) and which increase background particle number concentrations by up to 100 times, or 10^7 particles cm^{-3} (Zhu et al., 2011). Measurements have also shown aircraft emit secondary organic aerosol (SOA) precursors and the formation of SOA from those precursors can surpass primary aircraft PM emissions at idle (4% thrust), taxing (7% thrust), and approach (30% thrust) engine power settings (Miracolo et al., 2011). In this study, the focus is on the effects of aircraft emissions to fine particulate matter ($PM_{2.5}$) concentrations, and more specifically the SOA component of $PM_{2.5}$ at the Hartsfield-Jackson Atlanta International (ATL) Airport.

Organic compounds as a whole make up 20%–90% of aerosol mass in the lower troposphere (Kanakidou et al., 2005). Kroll and Seinfeld (2008) suggest that the pathways leading to the formation and evolution of SOA are likely the area with the largest uncertainties concerning organic aerosols in the atmosphere and while scientific understanding of SOA continues to evolve, it is believed there are still additional undiscovered as well as not yet fully understood precursors and pathways of SOA formation. Currently, the most widely accepted pathway is the oxidation of VOCs (primarily monoterpenes and aromatics) by free radicals—mainly the hydroxyl (•OH) radical, ozone (O_3), and nitrate (NO_3^-) radical—to form semi-volatile products of lower volatility which partition between the gas and particle phase (Kroll and Seinfeld, 2008).

Specific to aircraft emissions, only a limited number of studies have examined air quality impacts (Moussiopoulos et al., 1997; Brasseur et al., 1998; Pison and Menut, 2004; Carslaw et al., 2006) in an air quality model (AQM), none of which explicitly examine SOA production due to aircraft emissions. Woody et al. (2011) modeled air quality impacts to speciated $PM_{2.5}$ from aviation in a current and future year scenario from 99 major U.S. airports. Impacts to SOA near airports were insignificant, while inorganic species and elemental carbon comprised the majority of impacts. In fact, Woody et al. reported that despite aircraft emissions containing SOA precursors (e.g. xylene, toluene, benzene), they lowered total SOA concentrations at the Atlanta airport. Recent sampling and experimental results from Miracolo et al. (2011) suggest otherwise. Aircraft emissions from a CFM56-2B engine formed significant amounts of secondary PM after 3 h of photooxidation in a smog chamber (SOA at low engine power settings and sulfate aerosol at high engine power settings). The formation of relatively significant amounts of SOA at low engine power settings suggests that there are possible missing precursors from aircraft emission estimates currently used in AQMs.

At Atlanta, Unal et al. (2005) was one of the first to model aircraft impacts to $PM_{2.5}$, treating aircraft as line sources as opposed to the traditional point source treatment. However, only total $PM_{2.5}$ was considered. Subsequently, air quality and health impacts from commercial aircraft emissions were studied at the Atlanta, Chicago O'Hare, and Providence T.F. Green airports using a multiscale (36, 12, and 4-km) modeling approach (Arunachalam et al., 2011). A three-dimensional and realistic representation of aircraft emissions was developed using the EDMS2Inv tool (Baek et al., 2007), an interface between the FAA's Emissions and Dispersion Modeling System (EDMS) (Federal Register Notice, 1998), the required model for assessing air quality impacts from aviation sources in the U.S., and the Sparse Matrix Operator Kernel Emissions (SMOKE) model (Houyoux et al., 2000). Commercial aircraft emissions were included up to 3000 m and based on landing and take-off (LTO) cycles, which include startup, taxiing, queuing, takeoff, climb-out, and approach (Arunachalam et al., 2011). Aircraft emissions increased total $PM_{2.5}$ contributions overall both at and downwind of the three airports and grid resolutions considered. While the concentrations of most $PM_{2.5}$ species increased in the immediate vicinity of the ATL airport, nitrate and SOA concentrations decreased near the airport but increased downwind of it at the 36-km and 12-km resolutions (Arunachalam et al., 2011) agreeing with the SOA results reported by Woody et al. (2011) for ATL. At the 4-km grid resolution however, aircraft increased SOA concentrations both at and downwind of the ATL airport.

This work serves as an extension of the Arunachalam et al. (2011) study, and investigates the unexpected reduction of SOA concentrations near the airport due to aircraft emissions in the coarser grid resolutions despite SOA precursors contained in aircraft emissions. The overall objective is to determine model sensitivities of SOA concentrations from aircraft emissions at the ATL airport when using various model grid resolutions, and to determine the primary model processes responsible for this sensitivity.

2. Methodology

The Pennsylvania State University/NCAR mesoscale model (MM5) (Grell et al., 1994), SMOKE model, and Community Multiscale Air Quality (CMAQ) (Byun and Ching, 1999; Byun and Schere, 2006) v4.7 model were used to estimate the effects of aircraft emissions from the ATL airport on SOA. CMAQ treats PM formation through a trimodal approach (Binkowski and Roselle, 2003). $PM_{2.5}$ particles are represented by two lognormal distributions for the Aitken and accumulation modes; a third lognormal distribution represents coarse particles up to PM_{10}. CMAQ treats the following components of $PM_{2.5}$ explicitly in each of these modes: sulfate (ASO4), nitrate (ANO3), ammonium (ANH4), primary organic aerosol (POA), anthropogenic secondary organic aerosol (AORGA), biogenic secondary organic aerosol (AORGB) (total SOA = AORGA + AORGB), elemental carbon (AEC), and other unspeciated PM or crustal (A25). CMAQ v4.7 includes several updates in its treatment of $PM_{2.5}$ and SOA, including high/low-NO_x SOA pathways; the incremental evaluation of the updates is available elsewhere (Carlton et al., 2010; Foley et al., 2010).

Two emissions scenarios were considered: a base case with emissions estimated using the EPA's 2002 National Emissions Inventory (NEI) (EPA, 2007) and excluding the reported commercial aircraft emissions and a sensitivity case which included the base