Original article

Estimating ozone and secondary PM$_{2.5}$ impacts from hypothetical single source emissions in the central and eastern United States

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ABSTRACT

Secondary pollutant impacts from emissions of single sources may need to be assessed to satisfy a variety of regulatory requirements including the Clean Air Act New Source Review and Prevention of Significant Deterioration programs and the National Environmental Policy Act. In this work, single source impacts on O$_3$ and secondary PM$_{2.5}$ are estimated with annual 2011 photochemical grid model simulations where new hypothetical sources are added to the central and eastern United States with varying precursor emission rates and emission release heights. Impacts from these hypothetical sources are tracked with photochemical grid model source apportionment. Single source impacts on downwind 8-hr maximum O$_3$ tend to increase as emissions of NO$_X$ or VOC increase. Downwind impacts on PM$_{2.5}$ sulfate and nitrate also tend to increase as emissions of SO$_2$ and NO$_X$ increase. For all secondary pollutants, impacts from these hypothetical sources tend to decrease as distance from the source increases. However, peak impacts on O$_3$ and secondary PM$_{2.5}$ are not at the facility fence-line but typically within 50–100 km depending on the emissions rate, precursor pollutant, and emissions release point. Downwind impacts are not uniform directionally from these sources due to varying downwind availability of chemical reactants and prevailing meteorology. Peak impacts for O$_3$ (~15 ppb) and PM$_{2.5}$ sulfate (~8 μg/m^3) were within 50 km of these hypothetical sources and peak impacts for PM$_{2.5}$ nitrate (~1 μg/m^3) were within 125 km. The daily maximum 8-hr O$_3$ and maximum daily average PM$_{2.5}$ sulfate and nitrate ion impacts for the new hypothetical sources modeled here are generally consistent with those reported in literature. Additional assessments of single source impacts on secondary pollutants are still needed to provide a more comprehensive assessment of different source types and source environments.

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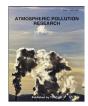
1. Introduction

Human exposure to fine particulate matter (PM$_{2.5}$, particles with aerodynamic diameter <2.5 μm) has been linked to a range of adverse health effects including cardiovascular and pulmonary disease (Künzli et al., 2005), lung cancer, and premature mortality (Pope III and Dockery, 2006). Exposure to ozone (O$_3$) has been linked to premature mortality (Bell et al., 2004) as well as aggravation of chronic health conditions such as bronchitis, emphysema and asthma (Devlin et al., 1997). Industrial facilities are well known

sources of primary PM$_{2.5}$ emissions and can also emit gaseous SO$_2$, NO$_X$, and VOC that can be precursors to secondarily formed O$_3$ and PM$_{2.5}$. The New Source Review (NSR) permitting program was established as part of the 1977 Clean Air Act Amendments. Nonattainment NSR permits are required for new major stationary sources or existing major stationary sources making a major modification in areas designated as nonattainment of the National Ambient Air Quality Standards (NAAQS). For areas attaining the NAAQS, sources may be required to obtain Prevention of Significant Deterioration (PSD) permits. Permits under PSD sometimes require a demonstration showing whether the new emission source will allow downwind areas to remain within the specific air quality guidelines.

The U.S. Environmental Protection Agency recently entered into a legal agreement to formulate guidance to assess O$_3$ and secondarily formed PM$_{2.5}$ impacts from single sources for permit programs

(U.S. Environmental Protection Agency, 2012). Air quality modeling methods to estimate the downwind impacts of primary pollutant emissions are well established through numerous dispersion modeling platforms that simulate plume dynamics and utilize meteorological data (http://www.epa.gov/ttn/scram/dispersion index.htm). However, the downwind impacts on O_3 and secondarily formed $PM_{2.5}$ from emissions of precursors is more complex to estimate because of the non-linear chemistry involved and the interaction of the plume with ambient levels of oxidants, neutralizing agents, and meteorology.

Attempts to estimate point source secondary pollutant impacts by including chemical mechanisms into plume models (Gaussian and Lagrangian) have had limited success because they often lack integration of a realistic ambient chemical environment. Regional scale Eulerian based chemical transport models (CTMs) like the Community Multiscale Air Quality Model (CMAQ; http://cmascenter.org/cmaq/) and the Comprehensive Air Quality Model with Extensions (CAMx; http://www.camx.com/) are capable of providing a realistic chemical and physical environment. Single source impacts have been estimated using photochemical grid models through various methods including brute force emissions adjustments (Cohan et al., 2005; Bergin et al., 2008; Zhou et al., 2012; Baker and Kelly, 2014; Kelly et al., 2015), decoupled direct method (DDM) (Cohan et al., 2005, 2006; Bergin et al., 2008; Baker and Kelly, 2014; Kelly et al., 2015), and emissions tagging source apportionment (Baker and Foley, 2011; Baker and Kelly, 2014). While these studies support the appropriateness of using photochemical grid models for single source secondary impact assessments, they do not provide a robust range of impacts covering a diverse set of sources, source environments, and time scales.

A primary motivation for a new systematic assessment of hypothetical sources using photochemical grid model source apportionment is to contribute more information about downwind single source secondary impacts to the existing literature. Here, new photochemical grid model simulations are presented to provide information about single source secondary pollutant impacts for a variety of locations, source emission rates, and plume release points. Photochemical grid model simulations for the entire year of 2011 have been completed with 24 hypothetical sources added to the central and eastern United States with varying emission rates and plume release heights. These hypothetical sources were systematically located near existing industrial point sources and spread out to cover a large geographic area. This is intended to provide a robust set of single source impacts that cover a variety of different chemical (e.g. VOC-to-NO_X ratio) and physical (e.g. meteorology and orography) regimes.

A review of published modeled single source secondary pollutant impacts is also presented to provide some context for the modeled impacts of the 24 hypothetical sources included in this analysis. Results from relevant photochemical model based single-source modeling studies published from 2005 through 2015 that report both the source emissions perturbation and corresponding change in downwind O_3 or $PM_{2.5}$ are summarized. Single source secondary impacts modeled as part of this study and those presented in existing literature provide some preliminary comparative information for future assessments done to support regulatory programs.

2. Methods

2.1. Air quality model simulations

Annual 2011 photochemical model simulations were performed for a domain covering the central and eastern United States with 12 km sized grid cells (Fig. 1). These simulations include a baseline simulation and 4 additional simulations each with 24 additional hypothetical sources added to the existing baseline emissions. Each hypothetical source was individually tracked with source apportionment for downwind contribution to O_3 and $PM_{2.5}$. All simulations were done using version 6.11 of the Comprehensive Air Quality Model with Extensions (CAMx) photochemical grid model (www.camx.com) (Baker and Scheff, 2007; Vizuete et al., 2008). This CAMx application includes ISO-RROPIA inorganic chemistry (Nenes et al., 1998), gas phase reactions based on the Carbon Bond (CB6r2) mechanism, and aqueous phase reactions (ENVIRON, 2014). Chemical boundary inflow is extracted from a photochemical model simulation for 2011 with a larger geographic domain covering the continental United States, northern Mexico, and southern Canada with 12 km sized grid cells. A total of 25 layers resolve the vertical atmosphere to 50 mb with thinner layers nearer the surface (layer one height is approximately 20 m). More details about the meteorological model simulation used to supply inputs to the emissions and CAMx model are available elsewhere (U.S. Environmental Protection Agency, 2014b).

Baseline emissions include anthropogenic sources based on version 2 of the 2011 National Emission Inventory (U.S. Environmental Protection Agency, 2014a) and biogenic sources estimated with the Biogenic Emission Inventory System version 3.6 (Carlton and Baker, 2011). Separate simulations were done where 24 hypothetical sources (see Fig. 1) were added to the baseline biogenic and anthropogenic emissions at varying emission rates of precursors and varying stack release characteristics (see Table S1) representing a "low" and "high" altitude plume release. Simulations where all 24 sources are emitting 500 and 1000 TPY of precursors (NO_X, VOC, and SO_2) were modeled with "low" plume release and additional simulations with sources emitting 1000 and 3000 TPY were modeled with "high" plume releases. These combinations result in a total of 4 different simulations that each included 24 hypothetical sources co-emitting NO_X, VOC, and SO_2 in addition to the baseline emissions (e.g. mobile, point, area sources). VOC (Table S2) and NO_X (90% NO and 10% NO_2) speciation for each of these hypothetical sources are based on average speciation profiles for non-EGU point sources.

The hypothetical source locations are intended to maximize distance between these sources while also being placed near existing industrial point sources (see Fig. 1). Most of these locations are rural or in close proximity to urban areas (Table S3). The contribution from each of these hypothetical sources to model predicted O_3 and $PM_{2.5}$ sulfate and nitrate ions were tracked using reactive tracers in the photochemical model (Kwok et al., 2013, 2015; ENVIRON, 2014). Ozone contributions were estimated using Ozone Source Apportionment Technology and $PM_{2.5}$ contributions using Particulate Source Apportionment Technology as implemented in CAMx (ENVIRON, 2014). All precursors are co-emitted but their impacts on $PM_{2.5}$ and O_3 are tracked separately (e.g. NO_X to O_3, VOC to O_3, etc.).

Photochemical models including CAMx have a Lagrangian based sub-grid plume treatment for representing plume chemistry and transport as an option for situations were grid volume dilution of emissions may not be desired (Karamchandani et al., 2011). Sub-grid plume treatment extensions in photochemical models typically solve for in-plume chemistry and use a set of physical and chemical criteria to determine when puff mass is merged back into the host model grid (Baker et al., 2014). Photochemical transport models have been shown to adequately capture single source impacts when applied using 4 km horizontal resolution when compared with downwind in-plume measurements (Zhou et al., 2012; Baker and Kelly, 2014). The horizontal grid resolution used here has been shown to provide similar single