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# Single source impacts estimated with photochemical model source sensitivity and apportionment approaches

Kirk R. Baker*, James T. Kelly

*U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, 109 TW Alexander Dr., Research Triangle Park, NC 27711, USA*

## HIGHLIGHTS

- Photochemical grid model can resolve single source impacts near-source and regionally.
- Source sensitivity and apportionment techniques capture single source impacts.
- Predict the primary and secondary average source impacts well at 4 km for this case.
- Capture downwind ozone production well (mean and peak) at 4 km for this case.
- Underpredict near-source ozone titration events and primary pollutant peaks at 4 km.

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## ABSTRACT

Some sources may need to estimate ozone and secondarily formed PM2.5 as part of the permit application process under the Clean Air Act New Source Review program. Photochemical grid models represent state-of-the-science gas- and particle-phase chemistry and provide a realistic chemical and physical environment for assessing changes in air quality resulting from changes in emissions. When using these tools for single source impact assessments, it is important to differentiate a single source impact from other emissions sources and to understand how well contemporary grid model applications capture near-source transport and chemistry. Here for the first time, both source apportionment and source sensitivity approaches (brute-force changes and high-order direct decoupled method) are used in a photochemical grid model to isolate impacts of a specific facility. These single source impacts are compared with in-plume measurements made as part of a well-characterized 1999 TVA Cumberland aircraft plume transect field study. The techniques were able to isolate the impacts of the TVA plume in a manner consistent with observations. The model predicted in-plume concentrations well when the observations were averaged to the grid scale, although peak concentrations of primary pollutants were generally underestimated near the source, possibly due to dilution in the 4-km grid cell.

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## 1. Introduction

Some facilities may be required to quantify emissions impacts of secondarily formed pollutants such as ozone ($O_3$) and particulate matter (PM) to satisfy permit review requirements of the New Source Review (NSR) program (U.S. Environmental Protection Agency, 2005). The United States Environmental Protection Agency (U.S. EPA) currently requires the use of AERMOD (a Gaussian steady-state dispersion model) to estimate the impacts of primarily emitted pollutants from single sources, but AERMOD does not simulate ozone or secondary $PM_{2.5}$ formation (Cimorelli et al., 2005). No specific model has been identified by U.S. EPA for near-field single source secondary impact assessments (U.S. Environmental Protection Agency, 2005). U.S. EPA recently granted a petition request from Sierra Club acknowledging the need to investigate methods for estimating the impacts of single sources on secondary pollutants (U.S. Environmental Protection Agency, 2014). Several air quality modeling tools could potentially be used for these assessments (ENVIRON, 2012), but very little information exists in the literature on the ability of such tools to simulate secondary pollutant formation from specific sources.

Some Lagrangian puff models include both ozone and PM chemistry and transport (e.g., SCICHEM). Impacts associated with the modeled source are clearly identifiable using such models since it is the only source in the model simulation (Karamchandani et al.,

* Corresponding author.
*E-mail address:* baker.kirk@epa.gov (K.R. Baker).

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2011). However, Lagrangian and Gaussian dispersion models require as input realistic information on the chemical and physical background environment to adequately characterize secondary formation that occurs at the edges of plumes. Photochemical grid models are advantageous in that they provide a realistic chemical and physical environment for single source secondary pollutant impacts because all sources are included in the simulation with multi-phase chemistry and pollutant transport. However, since photochemical grid models include all emissions sources, additional simulations or model extensions are needed to differentiate the impacts of a specific source from the others. These photochemical model based approaches generally fall into the categories of source apportionment and source sensitivity.

Photochemical model source apportionment allows specific emissions sources (or groups of sources) to be tracked from emission through transport, deposition processes, and chemical reactions to predict contributions to ozone and PM (Dunker et al., 2002a; Kwok et al., 2013; Wagstrom et al., 2008; Wang et al., 2009). Source sensitivity approaches provide information about how downwind modeled estimates of ozone and PM would change based on emissions changes in an identified source or group of sources. Source sensitivities can be estimated using the brute-force method or the direct decoupled method (DDM). A brute-force emissions change involves a second model simulation where emissions are modified compared to the original simulation. The difference in these simulations is an estimated impact of the source. An alternative is DDM, which internally tracks changes in air quality from the emissions of a particular source (Dunker et al., 2002b; Napelenok et al., 2006). Photochemical grid models have been used to estimate single source impacts on ozone using brute-force emissions adjustments (Bergin et al., 2008; Zhou et al., 2012) and DDM (Bergin et al., 2008). Single source impacts on secondarily formed $PM_{2.5}$ have been estimated using source apportionment (Baker and Foley, 2011).

While photochemical grid models have been applied with source sensitivity and apportionment methods to understand the air quality impacts from specific sources, fewer studies have demonstrated that photochemical models reasonably discern air quality impacts from a specific source (Zhou et al., 2012). Here, a plume measurement field study is used to explore the feasibility of using a photochemical grid model applied at fine grid resolution to distinguish the primary and secondary impacts of a single facility from other emissions sources. Measurements taken by aircraft at multiple downwind transects of a plume emitted from the TVA Cumberland power plant (Luria et al., 2001) are compared with photochemical model estimates. TVA Cumberland impacts are estimated with a photochemical grid model applied at fine grid scale (4-km grid cells) using multiple approaches to differentiate the facility from other sources. These approaches include source apportionment, brute-force emissions adjustment, and higher-order DDM (HDDM).

## 2. Methods

### 2.1. Field study transect measurements

Measurements of ozone ($O_3$), sulfur dioxide ($SO_2$), nitrogen oxides (NO and $NO_2$), and total reactive nitrogen compounds ($NO_Y$) were made at multiple transects downwind of the TVA Cumberland coal-fired power plant on July 6, 13, and 15 in 1999 (Luria et al., 2001, 2003). This facility is located in rural Tennessee approximately 80 km WNW of Nashville. Meteorological conditions on July 13 resulted in unusually low secondary chemical production in downwind transects compared to other flights during 1999 and previous field studies. Model estimated winds for July 13 resulted in no overlap between modeled and ambient-based plume estimates for transects on this day. The unfavorable meteorology on July 13 and potential measurement issues related to $NO_Y$ from the July 15 samples (Imhoff et al., 2001) leave the July 6 flights as the most appropriate sampling day for this evaluation. Although July 6 is the focus of this assessment, a comparison of model predictions and observations for July 13 and 15 is provided in the Supporting Information. Measurements along 12 plume traverses at four distances approximately 11–89 km downwind of the TVA Cumberland plant were taken on July 6 (see Table S1) in clear sky conditions near midday and late afternoon between 400 and 600 m in altitude (Luria et al., 2001, 2003). The aircraft transects through the TVA Cumberland plume only provide information about a specific trajectory through some portion of the plume and do not provide information about the total vertical and horizontal extent of the plume.

### 2.2. Photochemical grid model application

The Community Multiscale Air Quality model (CMAQ) version 5.01 (Byun and Schere, 2006) was applied to match the July 6, 1999, series of plume transect measurements made downwind of TVA Cumberland. The model simulation started on June 20, 1999 to minimize influence from initial conditions. CMAQ was applied with a domain centered over TVA Cumberland using 4 km grid cells that extend vertically from the surface layer (approximately 38 m depth) to 100 mbars using 34 layers (Fig. S1; Table S2). The 4-km model domain has spatially and time variant boundary conditions from coarser 36- (continental scale) and 12-km (regional scale) model domains. The prognostic Weather Research and Forecasting (WRF) model version 3.3 (Skamarock et al., 2008) was used to supply CMAQ gridded meteorological input data. The WRF configuration used for this application has been shown to reasonably predict mixing layer heights in other parts of the country during the summer season (Baker et al., 2013). Only model estimates from the 4-km domain are presented in this analysis.

CMAQ was configured with the Carbon-Bond 2005 gas phase chemical mechanism with toluene updates (Sarwar et al., 2011), aqueous reactions for sulfur and methylglyoxal (Sarwar et al., 2013; Walcek and Taylor, 1986), and AERO6 aerosol treatment which includes ISORROPA II inorganic chemistry (Fountoukis and Nenes, 2007) and semi-volatile organic partitioning between gas and aerosol phases (Carlton et al., 2010). Vertical mixing is based on the Asymmetric Convective Model (ACM2) hybrid local and non-local closure scheme (Pleim, 2007).

Emissions for the TVA Cumberland plant (Table S3) are based on day- and hour-specific Continuous Emissions Monitoring (CEM) database information (http://camddataandmaps.epa.gov/gdm/index.cfm?fuseaction=emissions.wizard). Emissions at other electrical generating units (EGUs) are based on annual total 2001 National Emission Inventory estimates allocated to specific days and hours in 1999 using CEM heat throughput data. All other anthropogenic emissions (including mobile, area, and non-EGU stationary point sources) are based on version 2 of the 2001 National Emission Inventory. Biogenic emissions are generated with Biogenic Emission Inventory System (BEIS) version 3.14 using temperature and solar radiation data output by the same WRF simulation used to generate input to CMAQ (Pierce et al., 1998). Biogenics estimated with BEIS compared well with field measurements taken during a similar time period (1998) and area: southern Illinois and Missouri (Carlton and Baker, 2011). Point sources are input to CMAQ using specific location coordinates and other emissions are generated at 4-km resolution.

For this analysis, CMAQ was applied using 3 different methods to isolate single source impacts: brute force zero out, DDM, and