



Interagency Workgroup on Air Quality Modeling Phase 3 Summary Report: Near-Field Single Source Secondary Impacts

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Single Source Secondary Impacts**

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Executive Summary

The Interagency Workgroup on Air Quality Modeling (IWAQM) was originally formed in 1991 to provide a forum for development of technically sound regional air quality models for regulatory assessments of pollutant source impacts on Federal Class I areas. The IWAQM process largely concluded in 1998 with the publication of the Interagency Workgroup on Air Quality Modeling (IWAQM) Phase 2 Summary Report and Recommendations for Modeling Long Range Transport Impacts (EPA-454/R-98-019) (U.S. Environmental Protection Agency, 1998). The IWAQM Phase 2 process provided a series of recommendations concerning the application of the CALPUFF model for use in long range transport (LRT) modeling and informed the promulgation of that model for such regulatory purposes in 2003. The IWAQM process was reinitiated in June 2013 to inform EPA's commitment to update Appendix W to address chemically reactive pollutants in near field and long range transport applications (U.S. Environmental Protection Agency, 2012a). This report provides information and recommendations from the "Phase 3" effort focused on near-field single source impacts of secondary pollutants.

This document describes chemical and physical processes important to the formation of ground-level O₃ and PM_{2.5} in the context of modeled near-field assessments to support permit review programs. Chemical transport models that characterize these processes include both Lagrangian which typically only have a single source included in the model and photochemical grid models which include some representation of all anthropogenic, biogenic, and geogenic sources. Modeling systems appropriate for the purposes of estimating single source near-field secondary impacts are described and recommendations are made with respect to the use of certain types of modeling systems for this type of application. Model evaluation is important to ensure that a particular system is fit for the purpose of estimating near-field single source secondary impacts. In addition to establishing a modeling system is generally appropriate for this purpose, project specific evaluations that compare model estimated meteorology and chemical estimates with measurements near the project source and key receptors is also an important model evaluation component.

An illustrative example is provided showing hypothetical single source impacts in two different urban areas: Atlanta and Detroit. A photochemical grid model was applied with baseline emissions and subsequent additional simulations where a new hypothetical source was included with a fixed precursor emission rate. The simulations with the additional hypothetical source are compared with the baseline simulation where the hypothetical source is not included (e.g. brute-force difference) and single source impacts are estimated for O₃ and secondary PM_{2.5} sulfate and nitrate. Downwind impacts from these hypothetical sources tend to increase as precursor emissions increase. Impacts tend to be highest within 15 km the source and decrease as distance from the source increases. However, there is variability in downwind impacts directionally from each of these sources due to differences in meteorology and available oxidants and neutralizing agents.

Finally, a review of existing research published between 2005 and 2015 relating single source precursor emissions and downwind impacts on O₃ and secondary PM_{2.5} is summarized. Downwind O₃ impacts from these studies show a general increase as NO_x emissions increase. Downwind impacts for O₃ and secondary PM_{2.5} in the studies reviewed tend to be largest nearest the source and decrease as distance from the source increases. Approaches for developing reduced-form or screening versions of more refined modeling tools such as photochemical transport models are also reviewed. At this time, it is not

clear that a robust reduced form model exists for either O_3 or secondary $PM_{2.5}$ for the purposes of assessing single source downwind impacts of these pollutants.

Table of Contents

1	Background: IWAQM Phase 3 process.....	6
2	Regulatory Motivation	6
3	Model Selection	7
3.1	Secondary Pollutant Formation: O ₃ and PM _{2.5}	7
3.2	Air Quality Models for Secondary Pollutants.....	7
3.2.1	Lagrangian models	8
3.2.2	Photochemical transport models.....	8
3.3	Recommendations for estimating single source O ₃ and secondary PM impacts using photochemical grid models	9
4	Model evaluation	10
4.1	Fit for Purpose Evaluations	10
4.2	Model evaluation: meteorology	11
4.3	Model evaluation: chemistry	11
4.4	Model performance evaluation data sources.....	12
5	Illustrative example of near-field single source impacts on O ₃ and PM _{2.5}	14
5.1	Model application approach for estimating these illustrative example single source impacts .	14
5.2	Illustrative single source impact results for O ₃ and PM _{2.5}	15
6	Review of published single source impacts on O ₃ and PM _{2.5}	19
6.1	Single source ozone impacts.....	19
6.2	Single source PM impacts	21
7	Reduced form approaches for estimating single source secondary impacts	22
7.1	Review of reduced form approaches for estimating O ₃ single source impacts.....	23
7.2	Review of reduced form approaches for estimating PM _{2.5} single source impacts	23
8	Acknowledgements.....	24
9	References	24

1 Background: IWAQM Phase 3 process

The Interagency Workgroup on Air Quality Modeling (IWAQM) was originally formed in 1991 to provide a forum for development of technically sound regional air quality models for regulatory assessments of pollutant source impacts on Federal Class I areas. Meetings were held with personnel from participating Federal agencies: the Environmental Protection Agency (EPA), the U.S. Forest Service (USFS), the U.S. Fish and Wildlife Service (USFWS), and the National Park Service (NPS). The original purpose was to review respective modeling programs, develop an organizational framework, and formulate reasonable objectives and plans for single source model applications. The IWAQM process largely concluded in 1998 with the publication of the Interagency Workgroup on Air Quality Modeling (IWAQM) Phase 2 Summary Report and Recommendations for Modeling Long Range Transport Impacts (EPA-454/R-98-019) (U.S. Environmental Protection Agency, 1998). The IWAQM Phase 2 report provided a series of recommendations concerning the application of the CALPUFF model for use in long range transport (LRT) modeling and informed the promulgation of that model for such regulatory purposes in 2003. Draft updates to the IWAQM Phase 2 report were released in 2009 to better reflect the state-of-the-practice of long range transport modeling techniques based on experience gained since the early 2000s.

The IWAQM process was reinitiated in June 2013 to inform EPA's commitment to update Appendix W to address chemically reactive pollutants in near field and long range transport applications (U.S. Environmental Protection Agency, 2012a). Comments received from the 10th Modeling Conference (March 2012) from stakeholders support this interagency collaborative effort to provide additional guidance for modeling single source impacts on secondarily formed pollutants in the near-field and for long range transport. Stakeholder comments also support the idea of this collaborative effort working in parallel with separate stakeholder efforts to further model development and evaluation.

This "Phase 3" effort includes the establishment of 2 separate working groups, one focused on long-range transport of primary and secondary pollutants and the other on near-field single source impacts of secondary pollutants. While many of the objectives are similar for each of these groups, the focus and regulatory end-points are different. The primary objectives of this phase of IWAQM are shown below.

It is expected the "Phase 3" effort will continue with future efforts related to reviewing and responding to comments given on the 2015 proposed changes to Appendix W related to single source impact assessments for O₃ and secondary PM_{2.5}. IWAQM3 near-field impacts team members (affiliated with U.S. Environmental Protection Agency unless noted otherwise) include James Kelly, George Bridgers, Andy Hawkins, Randall Robinson, Jaime Julian, Rebecca Matichuk, Robert Kotchenruther, Rynda Kay, and Richard Monteith. Additional participation was provided by Erik Snyder, Robert Elleman, and Bret Anderson (U.S. Department of Agriculture).

2 Regulatory Motivation

Pursuant to 40 CFR part 51.166 and 52.21, subsections (k)(1)(i) and (k)(1)(ii), new or modified sources emitting in significant amounts (see 40 CFR part 51.166 and 52.21, subsection (b)(23)(i)) are required to demonstrate that the source under review does not cause or contribute to a violation of any applicable National Ambient Air Quality Standards (NAAQS) ((k)(1)(i)) or maximum allowable increases over a baseline concentration ((k)(1)(ii)). The relevant permitting authority administers the National Ambient Air Quality Standards and increments component of the air quality analysis.

3 Model Selection

This section describes the types of air quality impacts that need to be assessed and the tools that are best suited for this purpose. For a variety of regulatory programs, impacts on secondary pollutants such as O₃ and PM_{2.5} need to be assessed at two spatial scales (near-source and long-range transport). It is important that modeling systems used for these assessments be fit for this purpose and be evaluated for skill in replicating meteorology and atmospheric chemical and physical processes that result in secondary pollutants, and deposition.

3.1 Secondary Pollutant Formation: O₃ and PM_{2.5}

PM and O₃ are closely related to each other in that they share common sources of emissions and are formed in the atmosphere from chemical reactions with similar precursors (U.S. Environmental Protection Agency, 2005). Air pollutants formed through chemical reactions in the atmosphere are referred to as secondary pollutants. For example, ground-level ozone (O₃) is predominantly a secondary pollutant formed through nonlinear photochemical reactions driven by emissions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight. Warm temperatures, clear skies (abundant levels of solar radiation), and stagnant air masses (low wind speeds) increase ozone formation potential (Seinfeld and Pandis, 2012).

In the case of particulate matter with aerodynamic diameter less than 2.5 μm (PM_{2.5} or fine PM), PM_{2.5} can be either primary (i.e. emitted directly from sources) or secondary (formed in the atmosphere). The fraction of PM_{2.5} which is primary versus secondary varies by location and season. In the United States, PM_{2.5} is dominated by a variety of chemical species: ammonium sulfate, ammonium nitrate, organic carbon (OC) mass, elemental carbon (EC), and other soil compounds and oxidized metals. PM_{2.5} elemental (black) carbon and soil dust are both directly emitted into the atmosphere from primary sources. Organic carbon particulate is directly emitted from primary sources but also has a secondary component formed by atmospheric reactions of VOC emissions. PM_{2.5} sulfate, nitrate, and ammonium ions are predominantly the result of chemical reactions of the oxidized products of sulfur dioxide (SO₂) and NO_x emissions and direct ammonia (NH₃) emissions (Seinfeld and Pandis, 2012).

3.2 Air Quality Models for Secondary Pollutants

Single source impacts on secondary pollution including ozone and PM_{2.5} are becoming increasingly important for facility permit reviews under the Prevention of Significant Deterioration (PSD) program as well as New Source Review (NSR) and other regulatory programs. Gaussian dispersion models such as AERMOD have been used to quantify the near-field (less than 50 km) impacts of primary PM_{2.5} emissions from new or modified sources (Perry et al., 2005; U.S. Environmental Protection Agency, 2005, 2014). However, these types of models cannot treat the important chemical and physical processes of ozone and secondary PM_{2.5}.

Chemical transport models treat atmospheric chemical and physical processes such as gas and particle chemistry, deposition, and transport. There are two types of chemical transport models which are differentiated based on a fixed frame of reference (Eulerian grid based) or a frame of reference that moves with parcels of air between the source and receptor point (Lagrangian) (McMurry et al., 2004). Photochemical grid models are three-dimensional grid-based models that treat chemical and physical

processes in each grid cell and use Eulerian diffusion and transport processes to move chemical species to other grid cells (McMurry et al., 2004). These types of models are appropriate for assessment of near-field and regional scale reactive pollutant impacts from specific sources (Baker and Foley, 2011; Baker and Kelly, 2014; Bergin et al., 2008; Zhou et al., 2012) or all sources (Chen et al., 2014; Russell, 2008; Tesche et al., 2006). Photochemical transport models have been used extensively to support State Implementation Plans and explore relationships between inputs and air quality impacts in the United States and beyond (Cai et al., 2011; Civerolo et al., 2010; Hogrefe et al., 2011).

3.2.1 Lagrangian models

Quantifying secondary pollutant formation requires simulating chemical reactions and thermodynamic partitioning in a realistic chemical and physical environment. Some Lagrangian models treat in-plume gas and particulate chemistry. These models require as input background fields of time and space varying oxidant concentrations, and in the case of PM_{2.5} also neutralizing agents such as ammonia, because important secondary impacts happen when plume edges start to interact with the surrounding chemical environment (Baker and Kelly, 2014; ENVIRON, 2012b). These oxidant and neutralizing agents are not routinely measured, but can be generated with a three dimensional photochemical transport model. Photochemical models simulate a more realistic chemical and physical environment for plume growth and chemical transformation (Baker and Kelly, 2014; Zhou et al., 2012), but simulations may sometimes be more resource intensive than Lagrangian or dispersion models.

3.2.2 Photochemical transport models

Publicly available and documented Eulerian photochemical grid models such as the Comprehensive Air Quality Model with Extensions (CAMx) (ENVIRON, 2014) and the Community Multiscale Air Quality (CMAQ) (Byun and Schere, 2006) model treat emissions, chemical transformation, transport, and deposition using time and space variant meteorology. These modeling systems include primarily emitted species and secondarily formed pollutants such as ozone and PM_{2.5} (Chen et al., 2014; Civerolo et al., 2010; Russell, 2008; Tesche et al., 2006). Even though single source emissions are injected into a grid volume, photochemical transport models have been shown to adequately capture single source impacts when compared with downwind in-plume measurements (Baker and Kelly, 2014; Zhou et al., 2012). Where set up appropriately for the purposes of assessing the contribution of single sources to primary and secondarily formed pollutants, photochemical grid models could be used with a variety of approaches to estimate these impacts. These approaches generally fall into the category of source sensitivity (how air quality changes due to changes in emissions) and source apportionment (how emissions contribute to air quality levels under modeled atmospheric conditions).

The simplest source sensitivity approach (brute-force change to emissions) would be to simulate 2 sets of conditions, one with all emissions and one with the source of interest removed from the simulation (Cohan and Napelenok, 2011). The difference between these simulations provides an estimate of the air quality change related to the change in emissions from the project source. Another source sensitivity approach to identify the impacts of single sources on changes in model predicted air quality is the decoupled direct method (DDM), which tracks the sensitivity of an emissions source through all chemical and physical processes in the modeling system (Dunker et al., 2002). Sensitivity coefficients relating source emissions to air quality are estimated during the model simulation and output at the resolution of the host model.

Some photochemical models have been instrumented with source apportionment, which tracks emissions from specific sources through chemical transformation, transport, and deposition processes to estimate a contribution to predicted air quality at downwind receptors (Kwok et al., 2015; Kwok et al., 2013). Source apportionment has been used to differentiate the contribution from single sources on model predicted ozone and PM_{2.5} (Baker and Foley, 2011; Baker and Kelly, 2014). DDM has also been used to estimate O₃ and PM_{2.5} impacts from specific sources (Baker and Kelly, 2014; Bergin et al., 2008; Kelly et al., 2015) as well as the simpler brute-force sensitivity approach (Baker and Kelly, 2014; Bergin et al., 2008; Kelly et al., 2015; Zhou et al., 2012). Limited comparison of single source impacts between models (Baker et al., 2013) and approaches to identify single source impacts (Baker and Kelly, 2014; Baker et al., 2013) show generally similar downwind spatial gradients and impacts.

Near-source in-plume aircraft based measurement field studies provide another approach for evaluating model estimates of (near-source) downwind transport and chemical impacts from single stationary point sources (ENVIRON, 2012b). Photochemical grid model source apportionment and source sensitivity simulation of a single source downwind impacts compare well against field study primary and secondary ambient measurements (e.g. O₃) made in Tennessee and Texas (ENVIRON, 2012b). This work indicates photochemical grid models and source apportionment and source sensitivity approaches provide meaningful estimates of single source impacts. However, additional evaluations are needed for longer time periods and more diverse environments, both physical and chemical, to generate broader confidence in these approaches for this purpose. In particular, it is important to ensure that adequate model performance is achieved in areas with complex terrain and meteorology.

3.3 Recommendations for estimating single source O₃ and secondary PM impacts using photochemical grid models

Photochemical transport models are suitable for estimating single source O₃ and secondary PM_{2.5} impacts since important physical and chemical processes related to the formation and transport of both are realistically treated. Source sensitivity and apportionment techniques implemented in photochemical grid models have evolved sufficiently and provide the opportunity for estimating potential secondary pollutant impacts from one or a small group of emission sources. Photochemical grid models using meteorology output from prognostic meteorological models have demonstrated skill in estimating source-receptor relationships in the near-field (Baker and Kelly, 2014; ENVIRON, 2012b) and over long distances (ENVIRON, 2012a).

In situations of close proximity between the source and receptor, a photochemical model instrumented with sub-grid plume treatment and sampling could potentially represent these relationships. However, the simplest approach to better representing the spatial gradient in source-receptor relationships when they are in close proximity would be to use smaller sized grid cells. Sub-grid plume treatment extensions in photochemical models typically solve for in-plume chemistry and use a set of physical and chemical criteria for determination of when puff mass is merged back into the host model grid (Baker et al., 2014). Sub-grid plume (puff) sampling or sub-grid puff merging with host grid cell estimates is necessary because inherently in this type of system (sub-grid plume treatment in a photochemical grid model) some of the source's impacts on air quality are resolved in puffs at the sub-grid scale and some has been resolved in the 3-dimensional grid space. Just extracting sub-grid plume information or just 3-dimensional model output would omit some of the source's contribution to air quality. In practice, some type of source apportionment or source sensitivity (e.g. brute-force difference) would be necessary to

track in the grid resolved source contribution in addition to sub-grid plume treatment to fully capture source contribution when using sub-grid plume treatment.

Given the complexities in fully determining project source impacts in both the grid model and sub-grid puffs (Baker et al., 2014) and the tendency for puffs to stay aloft compared to grid-resolved mass (Baker et al., 2014; Kelly et al., 2015) the use of sub-grid plume treatment for the purposes of estimating project source impacts for PSD/NSR would typically not be recommended. Additionally, these tools sometimes exacerbate numerical instability that infrequently might occur in inorganic and aqueous chemical reactions (Kelly et al., 2015).

4 Model evaluation

There are multiple components to model evaluation for the purposes of assessing long range transport of secondary pollutants for AQRVs. First, an alternative modeling system as defined in Appendix W must meet certain criteria for this purpose (Appendix W Section 3.2.2.e). One type of evaluation for this type of modeling system for this purpose is to show that the modeling system is theoretically fit for purpose. A second evaluation component involves comparison to ambient measurements to assess whether the modeling system and generated inputs are appropriate for a specific project application.

Since PM_{2.5} and O₃ impacts may be estimated for single sources as part of a permit review process, it is important that a modeling system be able to capture single source primary (e.g. precursors) and secondary impacts. Near-source in-plume measurements are useful to develop confidence that a modeling system captures secondarily formed pollutants from specific sources. These types of assessments are typically only done occasionally when a modeling system has notably changed from previous testing or has never been evaluated for this purpose. This type of assessment is discussed in more detail in section 4.1.

A second type of evaluation fulfills the need to determine whether inputs to the modeling system for a specific scenario are adequate for the specific conditions of the project impact assessment (Appendix W Section 3.2.2.e). This type of evaluation usually consists of comparing model predictions with observation data that coincides with the episode being modeling for a permit review assessment. One of the most important questions in an evaluation concerns whether the prognostic or diagnostic meteorological fields are adequate for their intended use in supporting the project model application demonstration. Sections 4.2 and 4.3 cover project specific evaluation approaches that develop confidence that a particular model application is appropriate for the project source and key downwind receptors. It is important to emphasize that a broad evaluation of a model platform's skill in estimating meteorology or chemical measurements may not sufficiently illustrate the appropriateness of that platform for specific projects that will be focused on a narrow subset of the larger set of model inputs and outputs. Therefore, broad model platform evaluations should be supplemented with focused evaluation and discussion of the appropriateness of model inputs for specific project assessments.

4.1 Fit for Purpose Evaluations

Near-source in-plume aircraft based measurement field studies provide another an opportunity for evaluating model estimates of (near-source) downwind transport and chemical impacts from single stationary point sources (ENVIRON, 2012b). Since single source impacts in the near field are assessed in each direction from a source at evenly distributed receptor locations model system skill in plume

placement is not emphasized. Ideally, these modeling systems will capture near-source plume placement to best match plume evolution with the surrounding heterogeneous chemical environment. Model system skill in capturing secondary impacts is important in near-field permit related assessments and in-plume field measurements provide the best opportunity for evaluating model skill in capturing secondary impacts from a specific source. Often when comparing modeled and measured in-plume pollutants the model impacts are shifted spatially to match the location of the measured plume meaning the comparison is paired in time but not space and therefore emphasizing secondary pollutant formation over plume placement.

Photochemical grid model source apportionment and source sensitivity simulation of a single source downwind impacts compare well against field study primary and secondary ambient measurements made in Tennessee and Texas (Baker and Kelly, 2014; ENVIRON, 2012b). This work indicates photochemical grid models and source apportionment and source sensitivity approaches provide meaningful estimates of single source impacts. However, additional evaluations are needed for longer time periods and more diverse environments to generate broader confidence in these approaches for this purpose.

4.2 Model evaluation: meteorology

It is important to determine whether and to what extent confidence may be placed in a prognostic meteorological model's output fields (e.g., wind, temperature, mixing ratio, diffusivity, clouds/precipitation, and radiation) that will be used as input to models. Currently there is no bright line for meteorological model performance and acceptability. There is valid concern that establishment of such criteria, unless accompanied with a careful evaluation process might lead to the misuse of such goals as is occasionally the case with the accuracy, bias, and error statistics recommended for judging model performance. In spite of this concern, there remains nonetheless the need for some benchmarks against which to compare new prognostic and diagnostic model simulations. A significant amount of information (e.g. model performance metrics) can be developed by following typical evaluation procedures that will enable quantitative comparison of the meteorological modeling to other contemporary applications and to judge its suitability for use in modeling studies.

Development of the requisite meteorological databases necessary for use of photochemical transport models should conform to recommendations outlined in Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, PM_{2.5}, and Regional Haze (EPA-454/B-07-002) (U.S. Environmental Protection Agency, 2007). Demonstration of the adequacy of prognostic or diagnostic meteorological fields can be established through appropriate diagnostic and statistical performance evaluations consistent with recommendations provided in the appropriate model guidance (U.S. Environmental Protection Agency, 2007).

4.3 Model evaluation: chemistry

An operational evaluation is used to assess how accurately the model predicts observed concentrations. Therefore, an operational evaluation can provide a benchmark for model performance and identify model limitations and uncertainties that require diagnostic evaluation for further model development/improvement. An operational evaluation for PM_{2.5} is similar to that for ozone. Some important differences are that PM_{2.5} consists of many components and is typically measured with a 24-hour averaging time. The individual components of PM_{2.5} should be evaluated individually. In fact, it is

more important to evaluate the components of PM_{2.5} than to evaluate total PM_{2.5} itself. Apparent “good performance” for total PM_{2.5} does not indicate whether modeled PM_{2.5} is predicted for “the right reasons” (the proper mix of components). If performance of the major components is good, then performance for total PM_{2.5} should also be good. Databases that contain ambient O₃, PM_{2.5}, and key precursors are noted in section 4.4. Section 4.4 is not intended to provide an exhaustive review of all ambient databases but provide an initial set of data that could be used for this purpose.

Regardless of the modeling system (e.g. photochemical transport or Lagrangian puff model) used to estimate secondary impacts of ozone and/or PM_{2.5}, model estimates should be compared to observation data to generate confidence that the modeling system is representative of the local and regional air quality. For ozone related projects, model estimates of ozone should be compared with observations in both time and space. For PM_{2.5}, model estimates of speciated PM_{2.5} components (such as sulfate ion, nitrate ion, etc) should be matched in time and space with observation data in the model domain. Model performance metrics comparing observations and predictions are often used to summarize model performance. These metrics include mean bias, mean error, fractional bias, fractional error, and correlation coefficient (Simon et al., 2012). There are no specific levels of any model performance metric that indicate “acceptable” model performance. Model performance metrics should be compared with similar contemporary applications to assess how well the model performs (Simon et al., 2012).

Accepted performance standards for speciated and total PM_{2.5} and ozone for photochemical models used in attainment demonstrations may not be applicable for single source assessments. Since the emissions and release parameters for the project source are well known, a direct connection between general photochemical model performance and the ability of the modeling system to characterize the impacts of the project source would be difficult to make. It is important that any potential approaches for photochemical model performance for the purposes of single source assessments for PSD and NSR use an approach that would be universally applicable to any single source modeling system, which includes the Lagrangian models described above.

4.4 Model performance evaluation data sources

Provided below is an overview of some of the various ambient air monitoring networks currently available that provide relevant data for model evaluation purposes. Network methods and procedures are subject to change annually due to systematic review and/or updates to the current monitoring network/program. Please note, there are other available monitoring networks which are not mentioned here and more details on the networks and measurements should be obtained from other sources.

AQS: The Air Quality System (AQS) is not an air quality monitoring network. However it is a repository of ambient air pollution data and related meteorological data collected by EPA, state, local and tribal air pollution control agencies from tens of thousands of monitors. AQS contains all the routine hourly gaseous pollutant data collected from State and Local Air Monitoring Stations (SLAMS) and National Air Monitoring Stations (NAMS) sites. SLAMS is a dynamic network of monitors for state and local directed monitoring objectives (e.g., control strategy development). A subset of the SLAMS network, the NAMS has an emphasis on urban and multi-source areas (i.e, areas of maximum concentrations and high population density). The AQS database includes criteria pollutant data (SO₂, NO₂, O₃, and PM_{2.5}) and speciation data of particulate matter (SO₄, NO₃, NH₄, EC, and OC), and meteorological data. The data

are measured and reported on an hourly or daily average basis. An overview of the AQS can be found at <http://www.epa.gov/ttn/airs/airsaqs/index.htm>.

IMPROVE: The Interagency Monitoring of PROtected Visual Environments (IMPROVE) network began in 1985 as a cooperative visibility monitoring effort between EPA, federal land management agencies, and state air agencies (IMPROVE, 2000). Data are collected at Class I areas across the United States mostly at National Parks, National Wilderness Areas, and other protected pristine areas. Currently, there are approximately 160 IMPROVE rural/remote sites that have complete annual PM_{2.5} mass and/or PM_{2.5} species data. The website to obtain IMPROVE documentation and/or data is <http://vista.cira.colostate.edu/improve/>.

STN: The Speciation Trends Network (STN) began operation in 1999 to provide nationally consistent speciated PM_{2.5} data for the assessment of trends at representative sites in urban areas in the U.S. The STN was established by regulation and is a companion network to the mass-based Federal Reference Method (FRM) network implemented in support of the PM_{2.5} NAAQS. As part of a routine monitoring program, the STN quantifies mass concentrations and PM_{2.5} constituents, including numerous trace elements, ions (sulfate, nitrate, sodium, potassium, ammonium), elemental carbon, and organic carbon. In addition, there are approximately 181 supplemental speciation sites which are part of the STN network and are SLAMS sites. The STN data at trends sites are collected 1 in every 3 days, whereas supplemental sites collect data either 1 in every 3 days or 1 in every 6 days. Comprehensive information on the STN and related speciation monitoring can be found at <http://www.epa.gov/ttn/amtic/specgen.html> and <http://www.epa.gov/aqspubl1/select.html>.

CASTNet: Established in 1987, the Clean Air Status and Trends Network (CASTNet) is a dry deposition monitoring network where data are collected and reported as weekly average data (U.S. EPA, 2002b). Relevant CASTNet data includes weekly samples of inorganic PM_{2.5} species and ground-level ozone. More information can be obtained through the CASTNet website at <http://www.epa.gov/castnet/>.

SEARCH: The South Eastern Aerosol Research and CHaracterization (SEARCH) monitoring network was established in 1998 and is a coordinated effort between the public and private sector to characterize the chemical and physical composition as well as the geographical distribution and long-term trends of PM_{2.5} in the Southeastern U.S. SEARCH data are collected and reported on an hourly/daily basis. Background information regarding standard measurement techniques/protocols and data retrieval can be found at <http://www.atmospheric-research.com/studies/SEARCH/index.html>.

NADP: Initiated in the late 1970s, the National Acid Deposition Program (NADP) monitoring network began as a cooperative program between federal and state agencies, universities, electric utilities, and other industries to determine geographical patterns and trends in precipitation chemistry in the U.S. NADP collects and reports wet deposition measurements as weekly average data (NADP, 2002). The network is now known as NADP/NTN (National Trends Network) and measures sulfate, nitrate, hydrogen ion (measure of acidity), ammonia, chloride, and base cations (calcium, magnesium, potassium). Detailed information regarding the NADP/NTN monitoring network can be found at <http://nadp.sws.uiuc.edu/>.

5 Illustrative example of near-field single source impacts on O₃ and PM_{2.5}

Photochemical grid models have been used with a variety of approaches to isolate secondary pollutant impacts from specific sources, including brute-force sensitivity (Baker and Kelly, 2014; Bergin et al., 2008; Kelly et al., 2015), direct decoupled method (DDM) (Baker and Kelly, 2014; Kelly et al., 2015), and source apportionment (Baker and Foley, 2011; Baker and Kelly, 2014). Here, downwind impacts of ozone and PM_{2.5} are described for hypothetical single sources placed in the Atlanta and Detroit metropolitan areas. These illustrative hypothetical single source impacts are based on Community Multiscale Air Quality (CMAQ) model simulations done for two different urban areas using 4 km sized grid cells. Impacts are estimated on ozone and PM_{2.5} from a hypothetical emissions source of SO₂, NO_x, and VOC using the brute-force sensitivity approach. The approach taken for this assessment is not intended to be a prescriptive approach recommended by this process or EPA.

5.1 Model application approach for estimating these illustrative example single source impacts

The Community Multiscale Air Quality Model (CMAQ) version 5.0.1 (www.cmaq-model.org) was applied for the entire year of 2007 to estimate PM_{2.5} and ozone. Aerosol chemistry is based on the AERO6 option that includes ISORROPIAII inorganic partitioning and chemistry (Fountoukis and Nenes, 2007), aqueous phase chemistry that includes sulfur and methylglyoxal oxidation (Sarwar et al., 2013), and organic aerosol partitioning (Carlton et al., 2010). Gas phase chemistry is represented with the Carbon-Bond 05 gas phase chemical mechanism with toluene updates (Sarwar et al., 2011).

Two separate model domains were used covering the Detroit and Atlanta metropolitan areas with 4 km sized grid cells (Figure 5-1; left panels). The vertical domain extends to 50 mb using 25 layers (surface layer height is ~20 m) with most resolution in the boundary layer to capture important diurnal variation in mixing height. The Weather Research and Forecasting model (WRF), Advanced Research WRF core (ARW) model version 3.3 (Skamarock et al., 2008) was applied with a horizontal grid resolution of 4 km and 35 vertical layers. Additional details regarding photochemical and meteorological model application and evaluation are provided elsewhere (U.S. Environmental Protection Agency, 2013a, b).

Anthropogenic and biogenic emissions are processed for CMAQ input using the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system (<http://www.cmascenter.org/smoke/>). Stationary point and area sources are based on version 2 of the 2008 National Emissions Inventory (NEI) (U.S. Environmental Protection Agency, 2012b). Mobile source emissions are day specific for the model simulation period based on data submitted to the National Emission Inventory and generated with SMOKE-MOVES (<http://cmasceneter.org/smoke/>). Hourly WRF estimated solar radiation and temperature are input to the Biogenic Emission Inventory System (BEIS) version 3.14 to generate emissions estimates of speciated VOC and nitric oxide (Carlton and Baker, 2011). Hourly boundary inflow from a coarser domain covering the continental United States with 12 km sized grid cells account for emissions from sources outside the 4 km model domain.

In addition to the full set of anthropogenic and biogenic emissions for this model domain, new hypothetical emissions sources are added to illustrate single source impacts on the Atlanta and Detroit areas. VOC (Table 5-1) and NO_x (90% NO and 10% NO₂) speciation are based on average speciation

profiles for non-EGU point sources in these areas. The location of the fictitious sources are shown in Figure 5-1. The same set of stack parameters were used for each fictitious source location: stack height 19 m, stack diameter 1.2 m, exit temperature 424 K, and exit velocity 14 m/s. Separate photochemical model simulations were done for each hypothetical source location and precursor emission rate: 100 tpy of VOC, 100 tpy of NO_x, 100 tpy of SO₂, 300 tpy of VOC, 300 tpy of NO_x and 300 tpy of SO₂. This resulted in a total of 6 simulations for 2 different hypothetical source locations or 12 total simulations with hypothetical sources in addition to 2 baseline simulations (1 for each area without any hypothetical source).

Table 5-1. Hypothetical source VOC speciation

Carbon Bond Specie	Fraction	Carbon Bond Specie	Fraction
ALD2	0.0152	MEOH	0.0054
ALDX	0.0155	NVOL	0.0008
ETH	0.0324	OLE	0.1143
ETHA	0.0094	PAR	0.4057
ETOH	0.0090	TERP	0.0170
FORM	0.0757	TOL	0.1148
IOLE	0.0088	UNR	0.1080
ISOP	0.0007	XYL	0.0674

5.2 Illustrative single source impact results for O₃ and PM_{2.5}

Annual maximum 24-hr average PM_{2.5} sulfate ion impacts are shown spatially in Figure 5-1 and by distance from the source in Figure 5-2 for hypothetical sources emitting 100 and 300 tpy of SO₂ in the Detroit and Atlanta areas. Impacts are generally highest nearest the source and decrease as distance from the source increases. However, visual examination of the spatial extent of maximum concentrations for each source shows that peak secondary impacts are not always coincident with the location of the emissions release. Occasional increases in downwind secondary impacts are generally related to single source precursor emissions reaching an area of increased oxidant availability, neutralizing agents, differences in terrain, mixing layer, or some combination of these influences.

The magnitudes and patterns of downwind impacts vary between these two areas and even within areas. This is likely due to differences in terrain features, available oxidants, and neutralizing chemicals such as ammonia. Different magnitudes and spatial patterns between these different source locations are more evident when looking at maximum 24-hr impacts compared to annual average. Maximum impacts tend to be dominated by conducive meteorology and nearby chemical environment on fewer days compared to the longer term average impacts, which is expected.

Figure 5-1. Annual maximum 24-hr PM_{2.5} sulfate ion impacts from a hypothetical source emitting 100 and 300 tpy of SO₂ at a location in Atlanta (top row) and Detroit (bottom row).

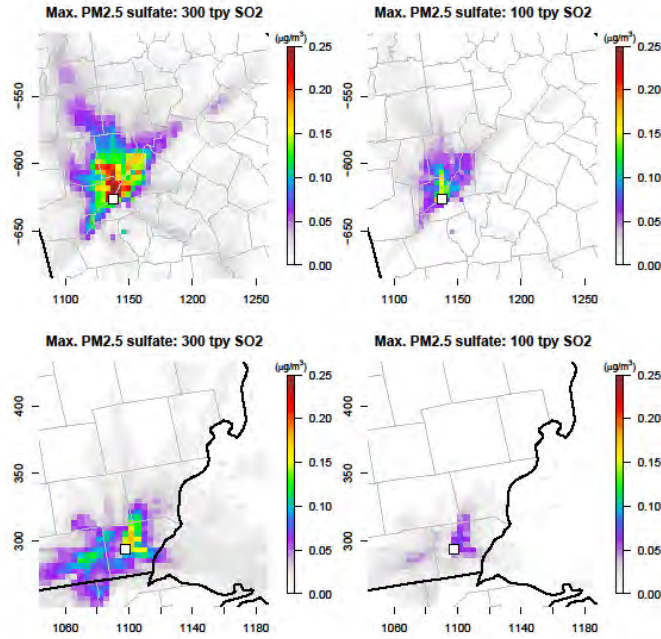
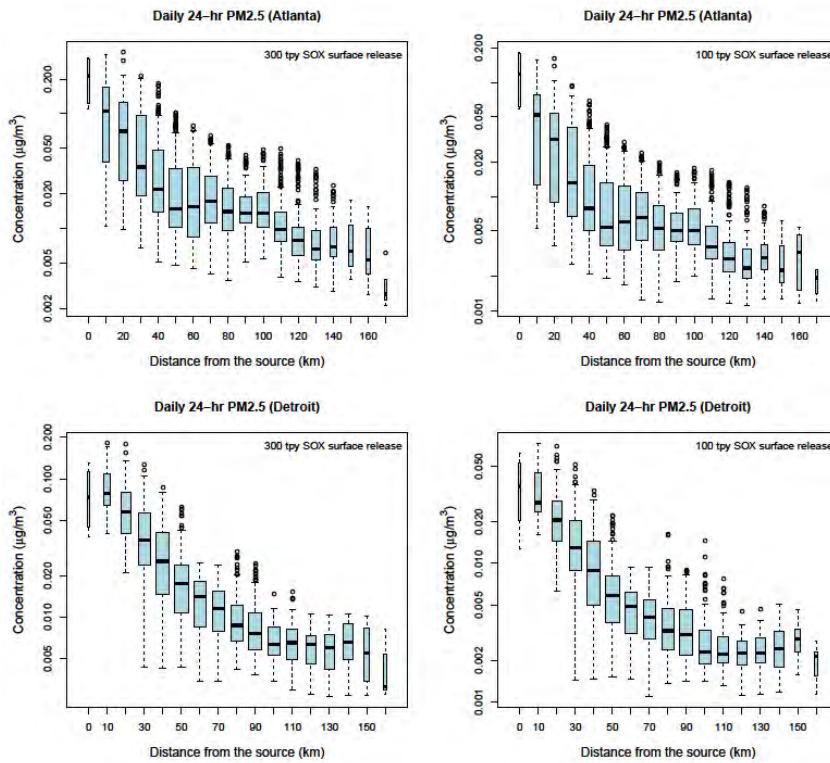


Figure 5-2. Maximum 24-hr PM_{2.5} sulfate ion impacts from a hypothetical source emitting 100 and 300 tpy of SO₂ at a location in Atlanta and Detroit by distance from the source. Results for Atlanta are shown on the top row and Detroit the bottom row.



Daily maximum 8-hr O₃ impacts over all days in the ozone season are shown as a function of distance from the hypothetical source emitting NO_x in Figure 5-3 and spatially in Figure 5-4. Similar information is provided in Figures 5-5 and 5-6 for hypothetical source emissions of VOC. Ozone impacts tend to be highest in grid cells adjacent to the hypothetical source and contributions generally decrease as distance from the source increases. Emissions of nitrogen oxides are concentrated enough in the 4 km grid cell containing the source that titration often dominates over production in that grid cell. For these particular urban areas and VOC emission mixtures, the hypothetical sources emitting NO_x emissions tended to form more O₃ than when emitting similar amounts of VOC emissions. Ozone impacts from the hypothetical source placed in Detroit shows notable impacts over Lake Erie. This is more prominent for the scenario of VOC emissions rather than NO_x emissions. It is possible that VOC from this hypothetical source encounters a favorable combination of available NO_x emissions from commercial marine sources and low boundary layer heights resulting in larger downwind O₃ contribution than in other directions from the hypothetical source.

Figure 5-3. Distribution of ozone season highest daily 8-hr maximum O₃ by distance from the hypothetical source NO_x emissions. Results for Atlanta are shown on the top row and Detroit the bottom row.

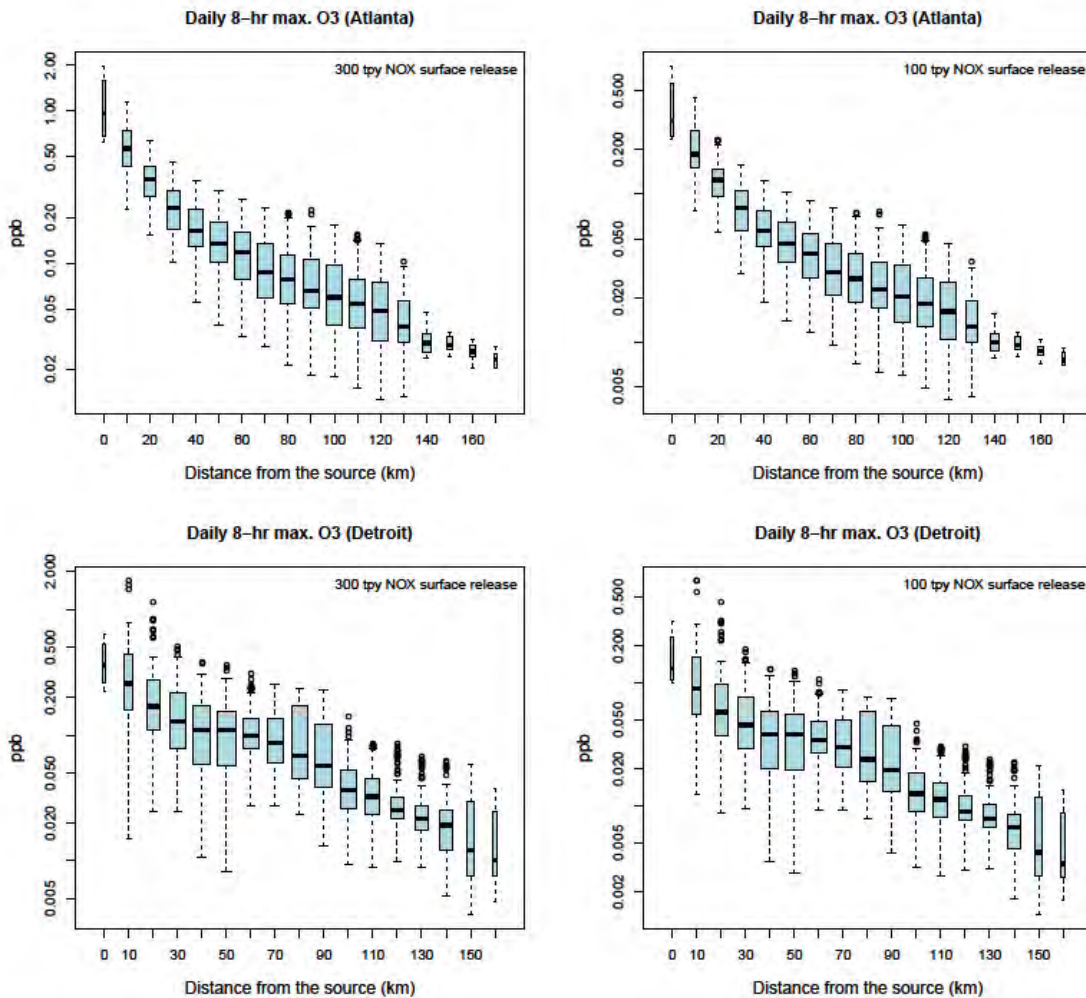


Figure 5-4. Spatial plot of ozone season highest daily 8-hr maximum O₃ from the hypothetical source NO_x emissions.

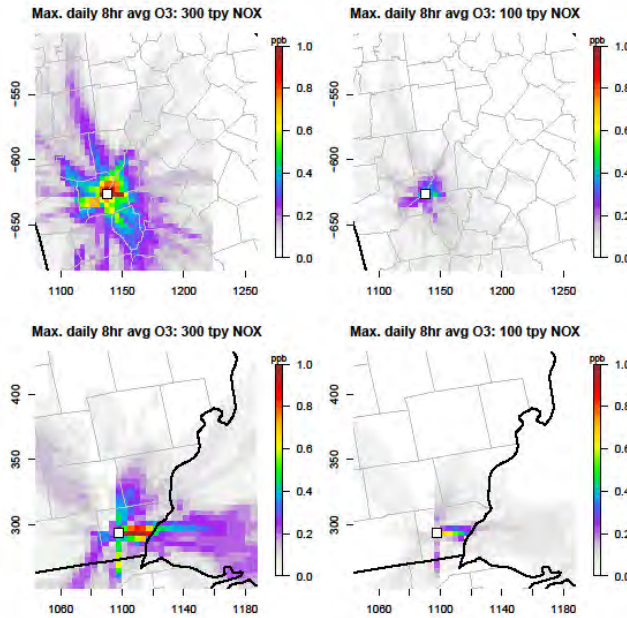


Figure 5-5. Distribution of ozone season highest daily 8-hr maximum O₃ by distance from the hypothetical source VOC emissions. Results for Atlanta on top row and Detroit the bottom row.

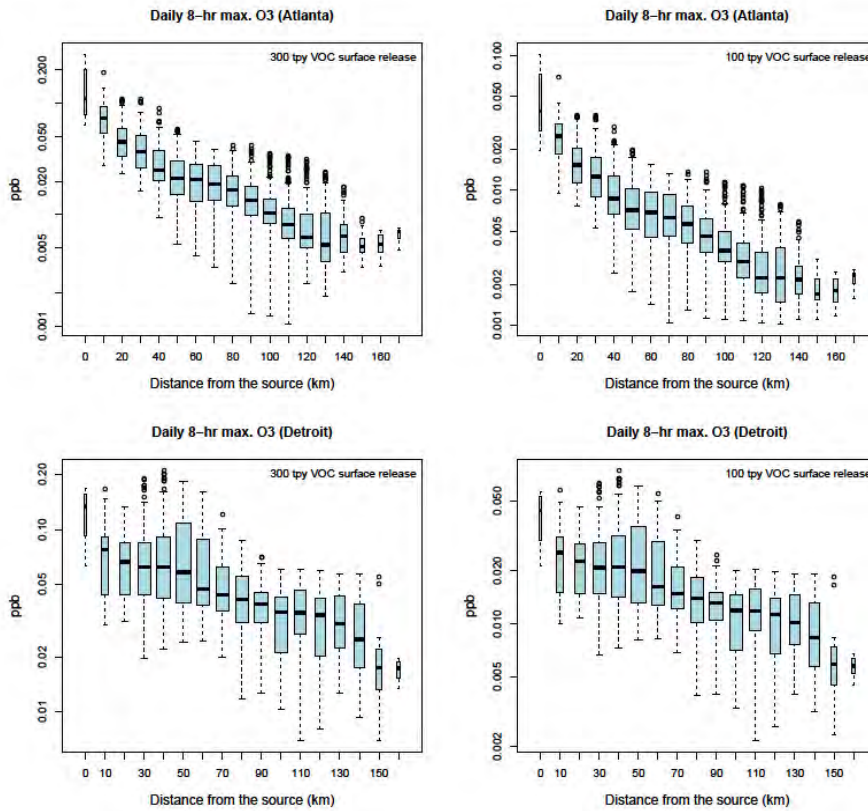
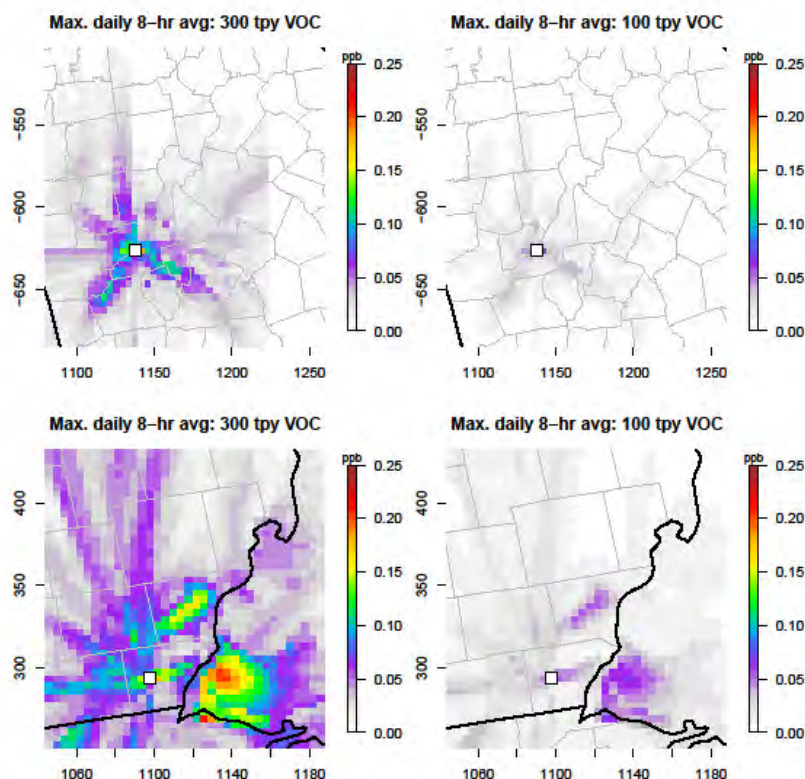


Figure 5-6. Spatial plot of ozone season highest daily 8-hr maximum O₃ from the hypothetical source VOC emissions.



6 Review of published single source impacts on O₃ and PM_{2.5}

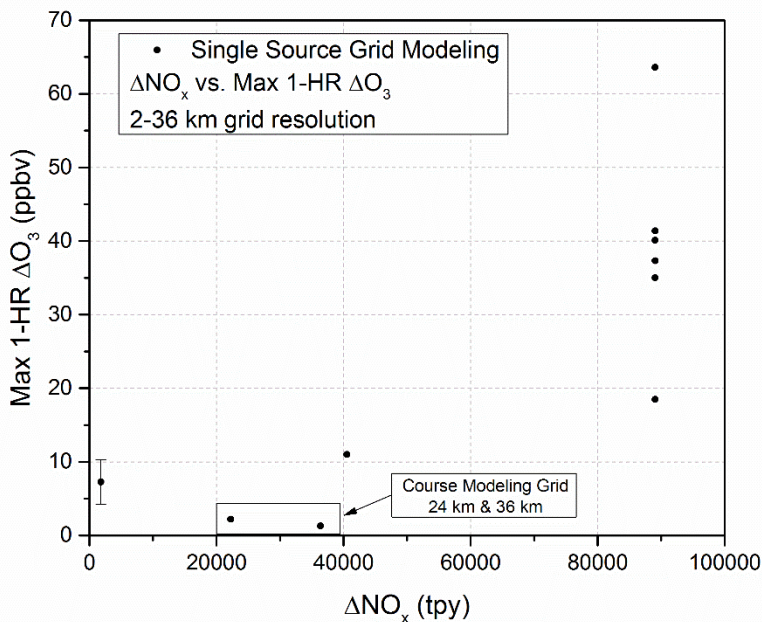
Published literature between 2005 and 2015 relating single source emissions with downwind O₃ and secondary PM_{2.5} impacts has been collected and summarized. This information reflects currently understood relationships between precursors and secondary pollutants and provides context for future model applications. The assessments reviewed here may not reflect every study done between 2005 and 2015. The primary criteria for inclusion in this report is a published study that presents both the emissions rate of a specific source and downwind impacts on O₃ or secondary PM_{2.5} from that same source. While the published studies provide useful information, 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.

6.1 Single source ozone impacts

Figure 6-1 presents the results from studies that quantified the 1 hr average maximum downwind O₃ from a single source NO_x emissions perturbation using photochemical grid models. Figure 6-2 shows the results from studies that quantified single source downwind 8 hr average O₃ impacts. For the studies

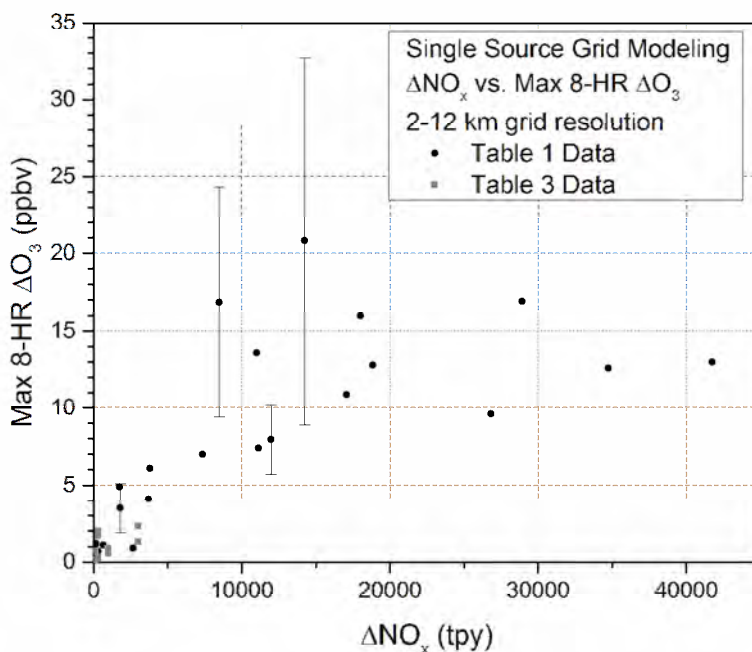
estimating downwind 1-hr O₃ contributions, there appears to be no clear pattern between NO_x emissions and maximum downwind O₃. A relationship is not evident even if results from studies with coarser model resolution (24 km, 36 km; N=2) are removed. However, Figure 6-2 shows a clearer relationship between NO_x emissions and 8-hr O₃. This may be due to the longer averaging time smoothing out some of the conditions that cause high variability in the 1 hr results. Another explanation for the notable relationship between emissions and secondary impacts for 8-hr O₃ is that most of the elements in Figure 6-2 are from a single publication that presented multiple source-impact relationships, meaning the consistent relationships are likely due to consistent model application and post-processing for each modeled source. Overall, peak 8-hr average O₃ impacts reported in literature range up to 33 ppb, which is a reported impact modeled from a large NO_x source (~14,000 tpy) over multiple high ozone episodes in the Louisiana/eastern Texas area (ENVIRON, 2005) using 4 km grid resolution. The results for single source VOC releases show very high variability in maximum downwind O₃ impacts due to large differences in ozone forming potential and OH reactivity of the VOC released.

Figure 6-1. Published single source NO_x emissions impacts on downwind hourly O₃ estimated using photochemical grid modeling.



Publications reporting results from observational field studies provide additional context to the model based studies for ozone production from point source emissions (Luria et al., 2003; Ryerson et al., 2001; Springston et al., 2005; Zhou et al., 2012). In these publications, in-plume observations related to specific sources were collected with aircraft and the sources examined were either power plants or petrochemical facilities. Many of these studies focused on quantifying ozone production efficiency from precursor emissions and typically did not provide key information to discern a relationship between source emissions and downwind O₃ impacts: a well-defined delta-O₃ (in plume O₃ minus an estimate of ambient background O₃) and facility precursor emissions. In some instances the study did not isolate impacts from a single facility plume but looked at some aggregate of nearby sources.

Figure 6-2. Published single source NO_x emissions impacts on downwind 8-hr average O₃ estimated using photochemical grid modeling.



One observation based study analyzed data from a field campaign that sampled plumes via helicopter downwind of the Tennessee Valley Authority Cumberland power plant on 4 summer days in 1998 and 1999 (Luria et al., 2003). These four days represented different NO_x emissions rates and meteorology. Three of the four days were conducive to ozone production, with one day having cooler temperatures where no positive delta-O₃ was observed at the farthest measured distance downwind. The three days with positive delta-O₃ reported the following maximum observed hourly delta-O₃ and corresponding NO_x emissions: 36 ppb O₃ (140,643 TPY NO_x), 39 ppb O₃ (71,697 TPY NO_x), and 24 ppb O₃ (115,681 TPY NO_x). While these NO_x emissions rates are generally higher than those of most of the modeling studies reported in Figure 6-1 (1-hr O₃), the measured change in O₃ appears to be in reasonable agreement with modeled estimates compiled from studies published in literature.

6.2 Single source PM impacts

A number of studies examine the effect of regional scale emissions on PM_{2.5} concentrations (Simon et al., 2012). Fewer studies have attempted to quantify the effects of single sources on downwind PM_{2.5} concentrations, and fewer still that report estimated secondary PM_{2.5} enhancements from these single sources (Baker and Foley, 2011; Baker and Kelly, 2014; ENVIRON, 2012a; National Association of Clean Air Agencies, 2011). Synthesizing the results of these limited studies is difficult due to differences in modeling (e.g., averaging timescales, PM_{2.5} species reported, etc), geographic area, emission profile and rates, stack height, near-source and regional NH₃ availability, and meteorology. Here, we summarize four studies which provide sufficient information on precursor emissions and secondary PM_{2.5} enhancements from single sources.

The NACAA Workgroup Final Report (National Association of Clean Air Agencies, 2011) provides a test case where PM_{2.5} impacts from different sources are assessed. The Minnesota case study presents predicted PM_{2.5} ammonium sulfate and ammonium nitrate from a full photochemical grid model simulation (CAMx) applied at 12 km grid resolution with sub-grid plume treatment and 200 m sub-grid plume sampling (National Association of Clean Air Agencies, 2011). Four individual stacks over Minnesota were chosen to reflect varying emission scenarios and background conditions. Emissions ranged from 400 to 13,000 tpy NO_x and 500 to 15,000 tpy SO₂, each with varying stack height. In these case study simulations all precursor emissions are co-emitted by each source (National Association of Clean Air Agencies, 2011). Over all cases, 24-hour maximum 98th percentile concentrations of secondary formation PM_{2.5} accounted for up to 1 µg/m³. These sources had stack release points well above ground level and had surface level peak impacts typically within 10 km of the source.

One published approach (Baker and Foley, 2011) use Particulate Matter Source Apportionment Technology (PSAT) applied with CAMx using 12 km grid resolution to estimate annual average secondarily formed PM_{2.5} sulfate and nitrate from precursor emissions. The study selects facilities representing the top 5% emitters of primary PM_{2.5}, NO_x, and SO_x from all stationary point sources located east of -97 longitude over midwest, mid-Atlantic, and southeast modeling domains (NO_x >7000 tpy, SO_x >20,000 tpy, and PM_{2.5} > 1100 tpy based on 2005 inventory). This study shows maximum annual average PM_{2.5} values of 0.385 µg/m³ and 0.018 µg/m³ for PM_{2.5} sulfate ion and PM_{2.5} nitrate ion respectively. Impacts were typically highest nearest the source and decrease as distance from the source increases (Baker and Foley, 2011).

Secondary PM_{2.5} was estimated for the Hunter EGU emitting 18,800 tpy of NO_x and 7,300 tpy of SO₂ in eastern Utah (ENVIRON, 2012). The CAMx model was applied with PSAT at 12 km resolution for an entire year. Predicted 24-hour (annual) maximum values were 3.44 µg/m³ (0.11 µg/m³) and 0.53 µg/m³ (0.05 µg/m³) of PM_{2.5} nitrate ion and PM_{2.5} sulfate ion respectively. Again, the highest secondary PM_{2.5} impacts tended to be closest to the source (ENVIRON, 2012).

A case-study examined the impact of a single EGU on downwind O₃ and PM_{2.5} sulfate ion using several photochemical grid model based source impact assessment approaches applied with the CMAQ model: brute force emission adjustments, DDM, and PSAT (Baker and Kelly, 2014). This case study examines emissions from the TVA Cumberland facility (48,000 tpy NO_x, 8,300 tpy SO₂) during a high pollution episode focusing on July 6, 1999. Maximum enhancements range up to 1.5 µg/m³ of PM_{2.5} sulfate ion. The different approaches for estimating single source impacts generally have similar spatial patterns.

7 Reduced form approaches for estimating single source secondary impacts

Predicting downwind secondary pollutant concentrations from point source emissions necessitates accounting for the interaction of the plume with ambient levels of oxidants, neutralizing agents, meteorology and potential nonlinear chemistry. State of the science approaches involve employing photochemical transport models like CMAQ and CAMx. There have been recent efforts to develop reduced form models that maintain the state of the science approach at their base, but provide a screening tool that is faster and cheaper to use. Screening tools that can provide rapid impact estimates for a wide range of emission source types (e.g. different emission loadings, stack heights, geographic locations) and can indicate whether more sophisticated approaches may provide credible information about the relationship between precursors and secondary impacts in some situations. Ideally, screening

level information would be an approximation of secondary impacts based on photochemical modeling of the single source, to take advantage of the state of the science treatment of multi-phase chemistry and deposition. This includes a realistic chemical and physical environment for single source impacts (Baker and Kelly, 2014).

7.1 Review of reduced form approaches for estimating O₃ single source impacts

In reviewing the available literature for this report, only one example of a reduced form O₃ modeling approach that incorporates a state of the science chemical transport model at its foundation has been identified. This approach was developed for the New South Wales (NSW) Greater Metropolitan Region in Australia (Yarwood et al., 2011). Briefly described, the Australian NSW approach involves a photochemical modeling analysis performed for several ozone seasons that includes hypothetical new emissions sources tracked using the higher order Decoupled Direct Method (HDDM) (Dunker et al., 2002) to calculate sensitivity coefficients for O₃ to the additional of NO_x and VOC emissions from the new hypothetical sources. The resulting O₃ sensitivity coefficients then allow O₃ impacts to be estimated for other NO_x and/or VOC sources within the same metropolitan area (Yarwood et al., 2011). While new sources or sources making modifications may not match the exact location, stack release characteristics, or emissions rates of the hypothetical sources in that analysis, options exist to pick the modeled source that best matches the new or modified source or even choose the highest impact from any of the modeled sources as a conservative approach.

7.2 Review of reduced form approaches for estimating PM_{2.5} single source impacts

Approaches for estimating secondary PM_{2.5} from single source emissions range from simple screening techniques to Gaussian based dispersion models and grid-based Eulerian photochemical transport models. The National Association of Clean Air Agencies recommended a multi-tiered approach for conducting air quality analysis of secondary PM_{2.5} ranging from initial screening tools to more refined analysis using complex photochemical modeling depending on the size and location of the source (National Association of Clean Air Agencies, 2011).

Screening techniques for secondary PM_{2.5} include the emissions divided by distance metric (Q/D) and the 100% conversion assumption. For the Q/D metric, allowable total emissions (Q) in tons per year are divided by distance to key receptors (D). This approach allows for a relative comparison of a variety of sources, but does not directly relate emission to concentrations for comparison to regulatory impact levels nor does it account for the variability in secondary PM_{2.5} formation. Under the 100% conversion assumption, NO_x and SO₂ emissions are assumed to convert entirely to ammonium nitrate and ammonium sulfate. A modeling study conducted with multiple case studies found that the 100% conversion approach was overly conservative, yielding physically unrealistic estimates with (National Association of Clean Air Agencies, 2011).

One approach (Baker and Foley, 2011) developed a nonlinear regression model relating large single source emissions over the eastern United States (N=99) to predicted annual average primary and secondary formed PM_{2.5} sulfate and nitrate as a function of emissions and distance. Individual regression models are developed for primary PM_{2.5}, PM_{2.5} sulfate ion, PM_{2.5} nitrate ion under favorable conditions, and PM_{2.5} nitrate ion under unfavorable conditions (Baker and Foley, 2011). This approach, a more physically realistic improvement to the Q/D approach, could be used to develop shorter-timescale (e.g., 24-hr), region and season specific regression models of PM_{2.5}. This nonlinear

regression approach has been applied to facilities in other countries to provide information about the downwind impacts of large point sources (Myllyvirta, 2014).

Inter-pollutant off-set ratios have been used to estimate secondary PM_{2.5} formation from single source SO₂ and NO_x emissions. In the case study of 4 EGUs in Minnesota described above, (National Association of Clean Air Agencies, 2011) estimate primary PM_{2.5} concentrations using AERMOD, then secondary concentrations were either added from CAMx results or using standard SO₂ and NO_x to primary PM_{2.5} offset ratios in EPA's New Source Review implementation rule for PM_{2.5} (73 FR 28321). The report concludes standard offset ratios lead to an over prediction of secondary PM_{2.5} and suggest a downward revision of offset ratios for the region. The report suggests new studies could be conducted to develop region, and season-specific offsets for use in the regulatory permitting context.

The photochemical grid model CAMx was used to examine the effects of distance, season, grid resolution, emission rate and stack height on predicted off-set ratios for the Port Washington coal-fired power plant in Georgia (Boylan and Kim, 2012). The study suggests maximum ratios for this facility are found near the source and vary most strongly with season and distance from the source. More recent work (Boylan and Kim, 2014) expanded on that study to investigate the spatial variability in seasonal offset ratios by modeling Port Washington emissions at 8 different locations in Georgia. Off-set ratios varied by up to a factor of 7 as a function of season or location, illustrating the sensitivity of secondary PM_{2.5} formation on meteorology and the local chemical environment.

Several of the studies reviewed presented promising techniques for developing regional scale reduced-form models to predict the impact of single sources on secondary pollutants. These approaches ranged from region specific ratios to more advanced regression models. Generalizing these techniques for areas not included in the original assessment is a difficult challenge but will be desired by many in the regulating and regulated communities.

8 Acknowledgements

The document includes contribution from Kirk Baker, Bob Kotchenruther, Rynda Kay, Bret Anderson, and Andy Hawkins. This document was reviewed by the IWAQM3-NFI group.

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United States
Environmental Protection
Agency

Office of Air Quality Planning and Standards
Air Quality Analysis Division
Research Triangle Park, NC

Publication No. EPA-454/P-15-002
July 2015
