Journal of the Air & Waste Management Association

ISSN: 1096-2247 (Print) 2162-2906 (Online) Journal homepage: http://www.tandfonline.com/loi/uawm20

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To cite this article: Kuo-Jen Liao & Xiangting Hou (2015) Optimization of multipollutant air quality management strategies: A case study for five cities in the United States, Journal of the Air & Waste Management Association, 65:6, 732-742, DOI: 10.1080/10962247.2015.1014073

To link to this article: http://dx.doi.org/10.1080/10962247.2015.1014073

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Accepted author version posted online: 18 Feb 2015.
Published online: 18 Feb 2015.

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TECHNICAL PAPER

Optimization of multipollutant air quality management strategies: A case study for five cities in the United States

Kuo-Jen Liao* and Xiangting Hou

Department of Environmental Engineering, Texas A&M University–Kingsville, Kingsville, TX, USA

**Please address correspondence to: Kuo-Jen Liao, Department of Environmental Engineering, Texas A&M University–Kingsville, 917 W. Ave. B - MSC 213, Kingsville, TX 78363, USA; e-mail: kuo-jen.liao@tamuk.edu*

Developing regional air quality management strategies is a difficult task because formation of air pollutants is interdependent and air quality at different locations may have different responses to emissions from common sources. We developed an optimization-based model, OPtimal integrated Emission Reduction Alternatives (OPERA), which allows for identifications of least-cost control strategies for attaining multipollutant air quality targets at multiple locations simultaneously. To implement OPERA, first, sensitivities of air quality to precursor emission changes are quantified. Second, cost functions of emission reductions are estimated using a cost analysis tool that includes a pool of available control measures. The third step is to determine desired reductions in concentrations of air pollutants. The last step is to identify the optimal control strategies by minimizing costs of emission controls using the sensitivities of air pollutants to emission changes, cost functions, and constraints for feasible emission reduction ratios. A case study that investigates ozone and $PM_{2.5}$ air quality in the summer of 2007 for five major cities in the eastern United States is presented in this paper. The results of the OPERA calculations show that reductions in regional NOx and VOC as well as local primary $PM_{2.5}$ emissions were more cost-effective than SO_2 controls for decreasing ozone and total $PM_{2.5}$ concentrations in the summer of 2007. This was because reductions in SO_2 emissions would only decrease $PM_{2.5}$ concentrations, and reductions in primary $PM_{2.5}$ emissions were more cost-effective than SO_2 emission controls.

Implications: We developed an optimization-based model, OPtimal integrated Emission Reduction Alternatives (OPERA), which allows for identification of least-cost emission control strategies for attaining multipollutant air quality targets at multiple locations simultaneously. A major strength of OPERA is its flexibility, which allows for changes in air quality regulations, involving agencies, study regions, and so on, to be readily incorporated. Overall, it has been demonstrated that OPERA is useful in developing least-cost emission control strategies for achieving multipollutant air quality targets at multiple locations simultaneously and could be useful for policymakers developing integrated air quality management plans.

Introduction

Formation of regional air pollution is affected by precursors emitted from multiple sources and regions, and it is typically difficult to identify effective mitigation strategies for secondary air pollutants because they share common precursors and their formation is interdependent (Meng et al., 1997). In some cases, reductions in concentrations of multiple pollutants are trade-offs because their responses to controls of common emissions are opposite (Liao et al., 2008). When attainment of air quality targets for multiple locations is considered, identification of effective control strategies becomes even more difficult since air quality at different locations may have different responses to emissions from common sources. In the traditional framework, iterative air quality simulations and control measure selections are usually required for developing control strategies that can simultaneously achieve prescribed air quality targets for multiple pollutants at multiple locations (Cohan et al., 2007). Since regional air quality management is a complicated issue, efficient approaches are desired to support air quality managers identifying optimal control strategies among a wide variety of potential combinations of control alternatives (Chow, 2010). In regional air quality management, the optimal control strategies may represent combinations of various emission control measures that maximize public welfare or minimize costs of emission reductions, meet social policy objectives, or provide combinations of such concerns. Wesson et al. (2010) evaluated the relative benefits of implementing a multipollutant, risk-based framework compared with the traditional single-pollutant approach used in developing a State Implementation Plan (SIP). The subsequent work of Fann et al. (2011) considered health benefits in the multipollutant, risk-based optimization framework, in contrast to the current study, which focuses solely on the attainment cost. Both studies focused on the Detroit, MI, metropolitan area, and the framework may need iterative selections of control strategies to achieve multipollutant, risk-based targets.

Two important indicators of regional air quality are daily maximum 8-hr average ozone (MDA8h O_3) and daily average $PM_{2.5}$

Journal of the Air & Waste Management Association, 65(6):732–742, 2015. Copyright © 2015 A&WMA. ISSN: 1096-2247 print
DOI: 10.1080/10962247.2015.1014073 Submitted October 5, 2014; final version submitted January 27, 2015; accepted January 27, 2015.
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(particulate matter with an aerodynamic diameter less than 2.5 μm). Both of those metrics have been found to be associated with adverse human health effects (Bell et al., 2006; Bernard et al., 2001; Lippmann, 1993; Poschl, 2005). In current air quality management practices, development of emission control strategies for attaining ozone and development of those for $PM_{2.5}$ standards are usually considered separately (Cohan et al., 2007; National Research Council, 2004, p. 130), although their formation is coupled via chemical interactions (Meng et al., 1997). Previous studies showed that reductions in anthropogenic nitrogen oxides (NOx) emissions can decrease ground-level $PM_{2.5}$ concentrations but may increase ozone concentrations in urban areas (Liao et al., 2008). Similarly, controls of anthropogenic volatile organic compound (VOC) emissions could also cause opposite responses for changes in ozone and $PM_{2.5}$ levels. Therefore, in some cases, simultaneous reductions in ozone and $PM_{2.5}$ levels by emission controls become difficult tasks. The objective of this study is to develop an innovative approach that allows identification of least-cost control strategies for achieving multipollutant air quality targets at multiple locations simultaneously without iterative air quality simulations and selection processes. Here we propose an optimization-based model, OPtimal integrated Emission Reduction Alternatives (OPERA), to approach least-cost control strategies for real-world problems: multiple-precursor controls for multipollutant attainment at multiple locations. This paper presents the development of OPERA and a case study for identifying least-cost emission control strategies for achieving ambient ozone and $PM_{2.5}$ air quality targets in single cities and multiple cities as a whole in the central and eastern United States.

Methods

Development of OPtimal integrated Emission Reduction Alternatives (OPERA)

The technique of mathematical programming has been applied to a wide variety of environmental problems (ReVelle, 2000). In air quality management, several mathematical programming methods, including deterministic and stochastic linear programming (LP) and nonlinear programming (NLP), have been pursued in order to identify optimal emission controls in sources for pollution mitigations in receptors and meet prescribed standards (Cooper et al., 1997; Greenberg, 1995). Previous studies mainly focused on emission controls to reduce concentrations of single air pollutants at single locations (Fu et al., 2006; Loughlin et al., 2000; Yang et al., 2009). In this study, we develop an optimization-based model, OPERA, formulated as an NLP, to identify least-cost control strategies for attaining prescribed multipollutant air quality targets at multiple locations simultaneously. The implementation of OPERA involves following four steps. First, relationships between ambient pollutants (e.g., ozone and $PM_{2.5}$) levels and precursor emission controls are quantified through regional air quality simulations and sensitivity analyses. Second, cost functions of emission reductions are developed using a cost analysis tool that includes a pool of available control measures for different emission sectors and regions. The third step is to determine required reductions in concentrations of air pollutants for areas of interest. The last step is to identify the optimal control strategies from a wide variety of combinations of control measures; this is done by minimizing costs of emission controls using the cost functions, sensitivities of air pollutants to emission changes, and constraints for feasible emission reduction ratios.

The cost functions for emission reductions developed using a cost analysis tool (i.e., AirControlNET, discussed in the next section) are used as the objective function in OPERA (eq 1). To formulate OPERA, relationships between emission reductions and air pollutant levels need to be determined. In this study, sensitivities, calculated using a regional air quality model (i.e., Community Multiscale Air Quality Model, discussed in the next section), are used to determine how ambient ozone and $PM_{2.5}$ concentrations respond to reductions in emissions of their precursors. Furthermore, the feasible ranges of emission reductions, which can be determined by an emission cost analysis tool (e.g., AirControlNET), need to be included in OPERA to constrain solutions.

To formulate OPERA for mitigating ozone and $PM_{2.5}$ air pollution, we define the following decision variables and parameters:

I	number of emission species
J	number of regions
K	number of cities
$\Delta\varepsilon_{ij}$	reduction ratios of emission species i from region j
$Cost_{ij}$	cost function of emission species i reductions from region j
$Cost_{primary\ PM,\ k}$	cost function of primary $PM_{2.5}$ reductions from city k
$S^{(1)}_{O3,ijk}$ and $S^{(1)}_{PM,ijk}$	first-order sensitivity of O_3 and $PM_{2.5}$ in city k to emission species i from region j
$S^{(2)}_{O3,ijk}$ and $S^{(2)}_{PM,ijk}$	second-order sensitivity of O_3 and $PM_{2.5}$ in city k to emission species i from region j
$C_{baseline\ O3,\ k}$	baseline O_3 concentration in city k
$C_{target\ O3,\ k}$	target O_3 concentration in city k
$C_{baseline\ PM,\ k}$	baseline $PM_{2.5}$ concentration in city k
$C_{target\ PM,\ k}$	target $PM_{2.5}$ concentration in city k
R_{ij}	maximum available reduction ratio for emission species i from region j
$R_{primary\ PM,\ k}$	maximum available reduction ratio for primary $PM_{2.5}$ emissions from city k

The complete mathematical form of OPERA is stated as follows:

$$\text{Minimize} \quad \sum_{i}^{I}\sum_{j}^{J} Cost_{ij}\left(\Delta\varepsilon_{ij}\right) + \sum_{k}^{K} Cost_{primary\ PM,k}\left(\Delta\varepsilon_{primary\ PM,k}\right) \tag{1}$$

subject to

$$\sum_{i}^{I}\sum_{j}^{J} \Delta\varepsilon_{ij} S^{(1)}_{O_3,ijk} + \frac{1}{2}\Delta\varepsilon^2_{ij} S^{(2)}_{O_3,ijk} \geq C_{\text{baseline } O_3,k} - C_{\text{target}O_3,k} \qquad k = 1, 2, \ldots K \tag{2}$$

$$\sum_{i}^{I}\sum_{j}^{J}\Delta\varepsilon_{ij}\ S^{(1)}_{PM,ijk}+\frac{1}{2}\Delta\varepsilon^2_{ij}S^{(2)}_{PM,ijk}+\Delta\varepsilon_{primary\ PM,k}$$
$$C_{primary\ PM,k}\geq C_{baseline\ PM,k}-C_{target\ PM,k}\quad k=1,2,\ldots K \tag{3}$$

$$0\leq\Delta\varepsilon_{ij}\leq R_{ij}\qquad i=1,2,\ldots I \text{ and } j=1,2,\ldots J \tag{4}$$

$$0\leq\Delta\varepsilon_{primary\ PM,k}\leq R_{primary\ PM,k}\qquad k=1,2,\ldots K \tag{5}$$

Equation 1 is the objective function that calculates the total cost of emission controls for achieving predetermined air quality targets. Equation 2 states that decreases in ozone concentrations, attributed to controls of regional precursor emissions, should be larger than or equal to desired reductions in ozone levels. Equation 3 indicates that decreases in $PM_{2.5}$ levels, attributed to controls of regional precursor emissions as well as local primary $PM_{2.5}$ emissions, should be larger than or equal to desired reductions in $PM_{2.5}$ levels. Equations 4 and 5 are inequality constraints and represent that the solutions for the least-cost emission control measures should be within feasible ranges of emission reductions. The goal of OPERA is to find solutions that minimize the emission control costs when prescribed air quality targets (eqs 2 and 3) and constraints of emission reduction ratios (eqs 4 and 5) are satisfied. The maximum available reduction ratio (R) (which is the upper limit of the feasible ranges of emission reductions) in eqs. 4 and 5 can be determined using a cost analysis tool (e.g., AirControlNET) by choosing different criteria (e.g., maximum allowed costs of emission controls) for assessing the emission control cost. Since the cost function is a combination of several nonlinear functions driven from regression analyses, the objective function in OPERA (eq 1) is nonlinear, and the optimal solutions are calculated using MATLAB with the Interior-Point algorithm (MathWorks, 2009). Several sets of initial points are fed at the beginning when solving OPERA, since a good set of initial points could facilitate the convergence of the solutions (i.e., the solutions with the minimal cost can be identified). The initial points (i.e., 10%, 20%, 30%, 40%, and 50% of emission reductions) were selected based on preliminary calculations using OPERA. It is noted that solutions may not be available if all the constraints cannot be satisfied simultaneously. For example, when background ozone levels are high, prescribed air quality targets may not be achieved because required magnitudes of emission controls could be out of feasible ranges of emission reductions.

A case study for developing least-cost air quality management strategies for east United States

In this section, we present a case study with a target to mitigate ozone and $PM_{2.5}$ air pollution for five cities in the central and eastern United States. We specifically examine concentrations of daily maximum 8-hr average (MDA8h) ozone and daily average $PM_{2.5}$ on August 2, 2007, which had high air pollutant levels in many cities of the eastern United States (Liao et al., 2014). Five cities, Atlanta, GA, Chicago, IL, Washington, DC, New York, NY, and Philadelphia, PA, are investigated in this case study because many of them were out of attainment of the ozone and $PM_{2.5}$ National Ambient Air Quality Standards (NAAQS) in 2007 (EPA, 2011).

Air quality modeling and sensitivity analyses. In this study, the Community Multiscale Air Quality Model (CMAQ) version 4.7.1 (Byun and Schere, 2006), with the high-order decoupled direct method (HDDM) (Cohan et al., 2005; Hakami et al., 2003), is applied to simulate ozone and $PM_{2.5}$ concentrations as well as their responses to changes in precursor emissions (i.e., sensitivities). We use the Carbon Bond 05 (CB-05) gas-phase chemical mechanism (Yarwood et al., 2005) and AERO5 aerosol module in the CMAQ simulations. In addition to HDDM, the adjoint and brute force methods can also be applied to CMAQ to model the sensitivities of air pollutants to emission changes. The differences among the three approaches are presented in several publications (e.g., Cohan et al., 2005; Napelenok et al., 2006; Hakami et al., 2007) and are not specifically discussed here. Results from the Weather Research and Forecasting (WRF) (Skamarock et al., 2008) and Sparse Matrix Operator Kernel Emissions (SMOKE) (Center for Environmental Modeling for Policy Development [CEMPD], 2004) models are applied to provide meteorological data and emissions, respectively, required for the CMAQ-HDDM modeling. The emissions inventory used in this study was the 2007 Mid-Atlantic Regional Air Management Association (MARAMA) Level 2 inventory, developed for Ozone Transport Commission (OTC) SIP screening modeling and released in 2010 (Vickers et al., 2011). A uniform grid of 12- by 12-km horizontal cells with 34 vertical layers is employed in the simulations. HDDM provides first- and second-order sensitivities of air pollutants to emission changes, and its results are capable to describe the nonlinear response of air pollutant formation to an up to ~50% reduction in precursor emissions. If responses of ambient ozone to more than 50% emission reductions need to be assessed, a multistage HDDM assessment should be used as employed by the EPA in its recent ozone risk assessment (EPA, 2014). Changes in ozone and $PM_{2.5}$ concentrations attributed to emission reductions can approximated as (Cohan et al., 2005)

$$\Delta C(E_i,E_j)\approx-\Delta\varepsilon_i S^{(1)}_i-\Delta\varepsilon_j S^{(1)}_j-\frac{1}{2}\Delta\varepsilon_i^2 S^{(2)}_{i,i}-\frac{1}{2}\Delta\varepsilon_j^2 S^{(2)}_{j,j}-\Delta\varepsilon_i\Delta\varepsilon_j S^{(2)}_{i,j} \tag{6}$$

and

$$S^{(1)}_i=E_i\left(\partial C/\partial E_i\right),\ S^{(1)}_j=E_j\left(\partial C/\partial E_j\right)$$
$$S^{(2)}_{i,i}=E_i^2\left(\partial^2 C/\partial E_i^2\right),\ S^{(2)}_{j,j}=E_j^2\left(\partial^2 C/\partial E_i^2\right),$$
$$S^{(2)}_{i,j}=E_iE_j\left(\partial^2 C/\partial E_i\partial E_j\right)$$

where ΔC is the change in pollutant concentrations. $S_i^{(1)}$ and $S_j^{(1)}$ are the first-order sensitivities of pollutant concentrations (C) to source emissions i (E_i) and j (E_j), respectively. $S_{i,i}^{(2)}$, $S_{j,j}^{(2)}$, and $S_{i,j}^{(2)}$ are the second-order sensitivities of pollutant concentrations to changes in precursor emissions. We consider three air pollutant precursors in this analysis: NOx, VOC, and sulfur dioxide (SO_2). NOx and VOC are important precursors for ground-level ozone and $PM_{2.5}$ formation, while SO_2, in addition to NOx and VOC, is an important precursor of secondary $PM_{2.5}$ (Poschl, 2005). In this study, first-order sensitivities of $PM_{2.5}$ to SO_2 emission changes, as well as second-order sensitivities of ozone and $PM_{2.5}$ to NOx and VOC emission changes, are included in the OPERA modeling. The second-order ozone sensitivities are considered to address nonlinear responses of ozone levels to emission reductions (Cohan et al., 2005). The sensitivities (i.e., first-order, secondary-order, and cross sensitivities) of ozone and $PM_{2.5}$ used in the case study are shown later, in Tables 1 and 2, respectively. Not all second-order and cross sensitivities (e.g., sensitivities of $PM_{2.5}$ to NOx and SO_2) are included in the case study, to make the results easier to interpret.

Because ground-level ozone and $PM_{2.5}$ could be affected by precursors emitted from local sources and more distant areas (Sillman, 1999), air pollution is recognized as a regional problem. Here, contributions of anthropogenic SO_2, NOx, and VOC, emitted from four regions (OTR, CENRAP, LADCO, and SEMAP) in the eastern United States (Figure 1), to MDA8h O_3 and daily average $PM_{2.5}$ concentrations in each of the five cities are estimated based on the results of the CMAQ-HDDM simulations. The assignment of the modeling domain to the four regions is based on the characteristics of precursor emissions and air pollutant formation. Furthermore, primary particulate matter could also be an important constitute of $PM_{2.5}$ in some urban areas (Kim et al., 2000a, 2000b), and its emissions mainly have local effects on ground-level $PM_{2.5}$ concentrations (Kim et al., 2002; Napelenok et al., 2007). Therefore, total $PM_{2.5}$ concentrations in each of the five cities are assumed to be affected by primary $PM_{2.5}$ (= primary elemental carbon [i.e., AECI, AECJ and A25J in CMAQ outputs] + primary organic carbon [i.e., AORPGAI and AORGPAJ in CMAQ outputs]) emitted from its Metropolitan Statistical Area (MSA), which includes the city center and surrounding counties (U.S. Census Bureau, 2012). Here, the effectiveness of reductions in local primary $PM_{2.5}$ emissions and regional SO_2, NOx, and VOC emissions for decreasing MDA8h O_3 and $PM_{2.5}$ levels is investigated for each of the cities and for multiple cities as a whole.

Figure 1. Air quality modeling domain, including four emission regions and five urban areas.

Development of cost functions for emission reductions. An EPA emission control analysis model, AirControlNET v4.1 (E.H. Pechan & Associates, 2006), is used to estimate the costs of emission reductions for different emission control measures (by species, region, cost per ton, etc.). AirControlNET mainly uses the EPA National Emission Inventory (NEI) for the year 1999 and relies on emission control efficiency and emission factor data provided in NEI to perform cost analyses. Results of AirControlNET provide mass of emissions reduced and associated annualized costs; that is, the costs of emission reductions are expressed as $Cost_{ij}(\Delta E_{ij})$, where ΔE_{ij} is the amount of emission reductions (e.g., ktons yr^{-1}) of species i from region j. ΔE_{ij} is determined when different criteria (i.e., maximum allowed costs of emission controls) are used in the AirControlNET analysis. Here, the cost functions are converted to be based on ratios of emission reductions (i.e., $Cost_{ij}(\Delta\varepsilon_{ij})$ in eq 7) since they are easier to incorporate in the OPERA formulations. Costs of reductions in anthropogenic precursor emissions do not increase linearly: Higher ratios of emission reductions are expected to be more expensive based on per ton reductions. For example, costs of per ton reductions increase significantly when anthropogenic VOC emissions are reduced more than about 60–70% (relative to total controllable emissions) (Figure 2). The total controllable emissions represent the sum of the emissions for which controls are applicable and used in the AirControlNET analyses (E.H. Pechan & Associates, 2006). Nonlinearity of cost functions is also found for regional SO_2 and NOx emission reductions (Figure 2). It is noted that AirControlNET is a somewhat dated tool and is used here since the EPA has not completed the release of a subsequent tool. A state-of-art cost analysis tool should be used in OPERA when it is available in the future.

Similar to reductions in SO_2, NOx, and VOC emissions, costs of per-ton reductions in primary $PM_{2.5}$ emitted from the five MSAs also increase nonlinearly with higher amount of emission reductions, and significant increases in the per-ton costs are found when reductions are more than about 40-50% (Figure 3). As such, the costs of emission reductions are fitted to an exponential or power function of absolute amount depending on species and regions. Given that using ratios of emission reductions in the optimization calculations could

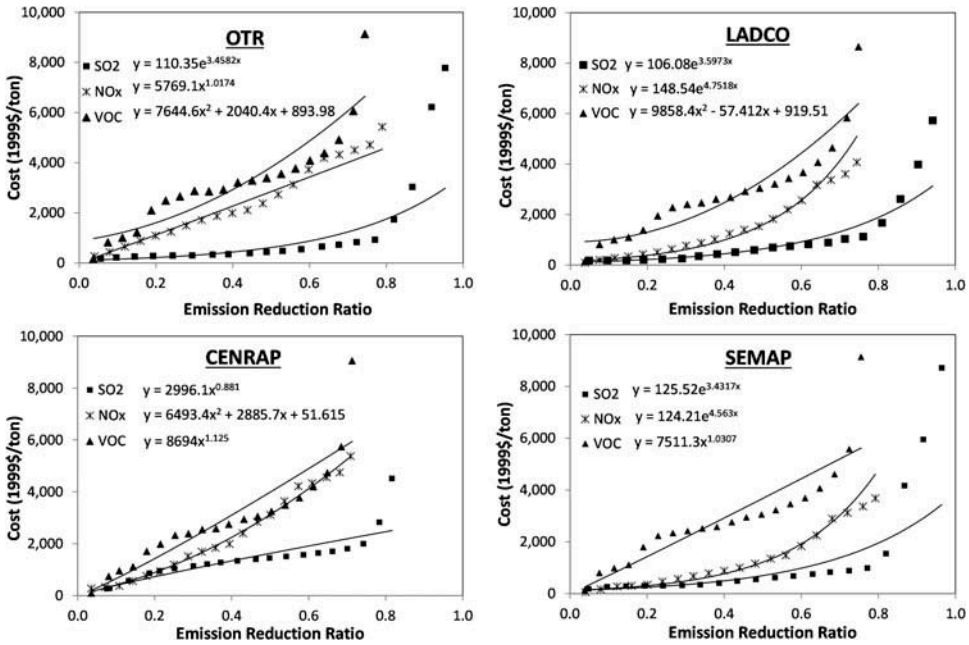

Figure 2. Per-ton cost (1999 dollars) of anthropogenic SO_2, NOx, and VOC emission reduction ratio (relative to total controllable emissions) from the four regions.

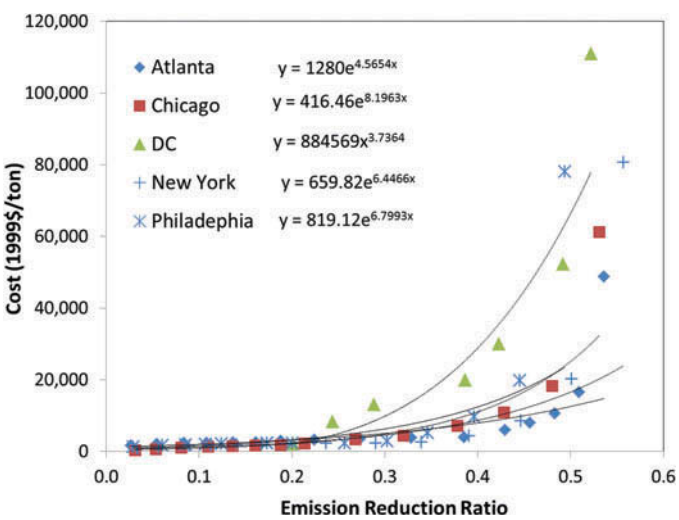

Figure 3. Per-ton cost (1999 dollars) of primary $PM_{2.5}$ emission reduction ratio (relative to total controllable emissions) from the five cities.

provide more explicit solutions, the cost functions of emission reductions have been rewritten in the following form:

$$Cost_{ij}(\Delta\varepsilon_{ij}) \text{ and } \Delta\varepsilon_{ij} = {}^{\Delta E_{ij}}/_{E_{ij}} \tag{7}$$

where $\Delta\varepsilon_{ij}$ is the reduction ratio of emission species i from region j (also used in eqs 1–4), ΔE_{ij} is the reduction in emissions (ktons yr^{-1}), and E_{ij} is the total controllable emission (ktons yr^{-1}). ΔE_{ij} and E_{ij} are calculated by AirControlNET when different criteria (i.e., maximum allowed costs of emission controls) are applied to the calculations. Finally, the costs become functions of emission reduction ratios, as presented in the form of eq 1, and they are applied in OPERA for calculating the least-cost emission control measures. It is important to note that the meaning of the reduction ratio in eq 7 is different from the rations in eqs 2–6 since AirControlNET only provides total controllable emissions rather than total emissions. Again, the issue should be solved when a state-of-art cost analysis tool is available in the future.

Emission controls on one species are expected to decrease emissions of other species from same sources in many cases (e.g., using cleaner fuels decreases NOx, VOC, and CO emissions from mobile sources). However, the emission reduction cost functions are estimated without considering synergy of the simultaneous emission reductions for different species from common sources because of limited sample size and degrees of freedom for regressions of the cost functions. Overall, the total costs of emission reductions are expressed as a linear summation of the costs of reductions in SO_2, NOx, and VOC emissions from the four regions and primary $PM_{2.5}$ emitted from the five cities (eqs 3 and 4). It is recognized that ignorance of co-benefits of emission reductions for multiple precursors could cause biases in the estimates of the cost functions and overestimations of control costs, but such biases are ignored in this study because our intention is to present a new method that allows for the identification of multipollutant air quality management strategies. A more rigorous investigation for the cost functions of emission reductions is an important task for decreasing the biases and should be further explored in the future.

For developments of control strategies for single cities, in total, 15 equations are used to constrain possible solutions for emission reductions. Among the 15 equations, 12 are used to constrain emission reduction ratios of the three precursors emitted from the four regions, one is used to constrain reduction ratios of primary

$PM_{2.5}$ emitted from the city, and two are used to define ozone and $PM_{2.5}$ reduction targets. When the five cities are considered simultaneously, 27 constraints are used in the calculations because 10 equations are used, instead of two, to define ozone and $PM_{2.5}$ reduction targets for the five cities, and five equations are applied to limit reduction ratios of primary $PM_{2.5}$ emitted from the five cities. The constraints on emission reduction ratios (i.e., R_{ij} in eq 4 and $R_{primary\ PM,\ k}$ in eq 5) used in the case study are determined by choosing maximum allowed emission control costs in AirControlNET for each of the four emission regions. Maximum costs of per-ton emission reductions can then be estimated based on the maximum allowed emission control costs and real emission reductions calculated by OPERA. The emission reduction constraints are not expected to affect results calculated by OPERA unless feasible solutions cannot be identified.

Results and Discussion

Baseline urban air quality and its sensitivity to emission controls

CMAQ-simulated daily maximum 8-hr average (MDA8h) ozone and 24-hr average $PM_{2.5}$ concentrations are compared against observations from various monitoring networks. The results show that ozone levels are slightly overestimated in Washington, DC (Liao et al., 2014), while $PM_{2.5}$ levels are underestimated in Atlanta, GA, and Philadelphia, PA, because CMAQ does not fully capture chemical formation mechanisms of secondary organic aerosols (SOAs) (Morris et al., 2006). However, the biases in the simulated air quality are not considered since the objective of this case study is to demonstrate how to apply OPERA to develop integrated ozone and $PM_{2.5}$ control strategies rather than to predict ambient ozone and $PM_{2.5}$ levels. In this case study, ozone and $PM_{2.5}$ air quality on August 2, 2007, is investigated due to high air pollutant levels on that day (Liao et al., 2014). Simulated MDA8h O_3 levels ranged from 61.7 (Chicago) to 91.8 (New York) parts per billion (ppb) on August 2, 2007, for the five cities (Table 1). Daily-averaged $PM_{2.5}$ levels are simulated to range from 11.7 (Washington, DC) to 38.8 (New York, NY) $\mu g/m^3$ on the same day. Primary $PM_{2.5}$ levels in the five cities are predicted to range from about 3.8 (Washington, DC) to 10.5 (New York, NY) $\mu g/m^3$ and contributed up to about one third of the total $PM_{2.5}$ levels in the five cities (Table 2). In general, the results of the CMAQ-HDDM sensitivity analysis show that MDA8h O_3 levels in the five cities were mainly affected by emissions from within the regions they are located. For example, NOx emissions from the OTR region had the most significant contribution to MDA8h O_3 concentrations in Washington, DC. On the other hand, reductions in NOx emissions from OTR could increase MDA8h O_3 concentrations in New York, NY, due to negative ozone sensitivities to NOx emission reductions (Table 1). They could be attributed to high NOx emissions from local sources and a VOC-limited ozone formation regime on August 2, 2007. Precursors emitted from distant regions resulting from transport of pollutants could affect ozone levels in the cities examined. In addition to Washington, DC, and Philadelphia, PA (located in the OTR region), anthropogenic NOx emissions from the OTR region are simulated to contribute to MDA8h ozone formation (~18 ppbv) in Atlanta, GA, which is located in the SEMAP region (Table 1). Sensitivities of MDA8h O_3 and daily-averaged $PM_{2.5}$ to emission changes on another polluted day (i.e., August 4, 2007) are presented in the appendix (Tables A.1 and A.2), and the results of MDA8h O_3 sensitivities in New York, NY, to NOx emissions from OTR were quite different from those on August 2. This implies that the sensitivities could vary significantly from day to day. However, the daily variation in the sensitivities is not specifically examined here since the objective of this case study is to illustrate OPERA.

Least-cost ozone and $PM_{2.5}$ air quality management strategies for single cities

NAAQS for 3-year averages of yearly fourth highest MDA8h ozone and daily average $PM_{2.5}$ are 75 ppb and 35 $\mu g/m^3$, respectively. However, only the baseline $PM_{2.5}$ concentration in New York, NY, was above 35 $\mu g/m^3$ during the study episode. To include more cities in the OPERA calculations, least-cost control strategies for achieving 75 ppb MDA8h ozone and 25 $\mu g/m^3$ daily $PM_{2.5}$ concentrations on August 2, 2007, are investigated for each of the five cities.

Atlanta, GA. Based on the results of the OPERA calculation, reductions in anthropogenic NOx emissions from the OTR (~16%), LADCO (~2%), and SEMAP (~39%) regions and in VOC emissions from SEMAP (~3%) were the least-cost control strategies for achieving the ozone and $PM_{2.5}$ air quality targets in Atlanta. The least cost of the emission reduction was about $1.3 billion (1999$) (Table 3). Decreases in NOx emissions from the OTR and SEMAP regions were cost-effective for achieving the prescribed air pollutant levels because they were important precursors for peak ozone formation on August 2, 2007, in Atlanta (Table 1). On the other hand, controls of SO_2 or local primary $PM_{2.5}$ emissions were not cost-effective for achieving the prescribed air quality targets in Atlanta; this was because such reductions would only decrease $PM_{2.5}$ levels that were already below the $PM_{2.5}$ target.

Chicago, IL. For ambient air quality on August 2, 2007, in Chicago, anthropogenic NOx and VOC emissions from the LADCO region were the most important contributors to ozone formation (Table 1). In addition to local primary $PM_{2.5}$ emissions, anthropogenic SO_2 and NOx emissions from LADCO were important contributors (about 6 $\mu g/m^3$ in total) to $PM_{2.5}$ levels in Chicago (Table 2). The results of OPERA calculations show that reductions in primary $PM_{2.5}$ emissions from the Chicago MSA (~14%) were the most cost-effective control measure for reducing the baseline daily average $PM_{2.5}$ concentration by 1.3 $\mu g/m^3$ (from 26.3 $\mu g/m^3$ to 25 $\mu g/m^3$) and achieving the prescribed air quality targets in Chicago. Reductions in NOx and VOC emissions were not required since the baseline MDA8h ozone level in Chicago was below

Table 1. Daily maximum 8-hr average (MDA8h) ozone concentrations (ppb) as well as first, second, and cross sensitivities of MDA8h ozone to NOx and VOC emissions on August 2, 2007

Region	Sensitivity/MDA8h O_3	Atlanta 84.8	Chicago 61.7	D.C. 85.6	New York 91.8	Philadelphia 84.4
OTR	1st NOx	12.20	0.22	55.30	–14.20	26.10
	1st VOC	0.04	0.04	–0.32	16.20	5.00
	2nd NOx	–11.60	–0.07	–39.70	–22.50	–50.20
	2nd VOC	–0.88	–0.01	–0.42	0.55	–1.53
	2nd NOx_VOC	2.40	0.01	2.60	–1.03	6.70
LADCO	1st NOx	0.42	5.20	0.52	–0.02	0.72
	1st VOC	0.09	3.60	–0.06	0.02	–0.02
	2nd NOx	–0.11	–19.80	–0.08	0.02	–0.09
	2nd VOC	–0.02	–0.59	~0	~0	~0
	2nd NOx_VOC	0.06	2.30	0.01	~0	0.01
CENRAP	1st NOx	0.16	0.57	0.30	0.01	0.33
	1st VOC	0.01	0.01	–0.01	~0	0.01
	2nd NOx	–0.03	–0.07	–0.04	~0	–0.04
	2nd VOC	~0	~0	~0	~0	~0
	2nd NOx_VOC	0.01	0.01	0.01	~0	0.01
SEMAP	1st NOx	26.80	0.02	0.09	~0	~0
	1st VOC	1.39	0.01	~0	~0	~0
	2nd NOx	–32.90	–0.01	–0.01	~0	~0
	2nd VOC	–0.19	~0	~0	~0	~0
	2nd NOx_VOC	1.89	~0	~0	~0	~0

the prescribed ozone air quality target. The least cost of the emission reduction was $253 million (1999 dollars).

Washington, DC. To achieve the ozone and $PM_{2.5}$ air quality targets in Washington, DC, reducing NOx emissions from the OTR region by ~21% was the least-cost control strategy (Table 3). Such a reduction would decrease MDA8h ozone concentrations from 85.6 to 75ppb in Washington, DC. On the other hand, reducing VOC emissions would not be effective for improving the ozone air quality since ozone formation in Washington, DC, was dominated by NOx emissions. Reductions in SO_2 emissions from the four regions or primary $PM_{2.5}$ from the MSA of Washington, DC, would not be necessary since the baseline $PM_{2.5}$ concentration (i.e., 11.7 μg/m^3) in Washington, DC, was below the $PM_{2.5}$ air quality target (i.e., 25 μg/m^3). The least cost of the emission reduction for achieving the prescribed air quality targets in Washington, DC, was $838 million (1999 dollars).

New York, NY. To achieve the prescribed ozone and $PM_{2.5}$ air quality targets on August 2, 2007, in New York, the baseline MDA8h ozone and daily-average $PM_{2.5}$ concentrations would need to be reduced by 16.8 ppb and 13.8 μg/m^3, respectively (Table 3). However, the results of the OPERA calculation show that there is no solution found within the constraints of emission reduction ratios. In other words, it was infeasible to achieve the prescribed ozone and $PM_{2.5}$ concentrations simultaneously in New York; this was mainly because of the negative sensitivities of ambient ozone concentrations in New York to NOx emissions from the OTR region. High background ozone levels and/or significant amount of long-range transported $PM_{2.5}$ could also make the air quality target more difficult to achieve. Given that the objective of this paper is to present the development and applicability of the new method for developing multipollutant air quality management strategies instead of predicting real air pollutant levels, examinations of background air pollutant levels and transported $PM_{2.5}$ for the simulated episode and cities are omitted.

Philadelphia, PA. To achieve the ozone and $PM_{2.5}$ air quality targets simultaneously on August 2, 2007, in Philadelphia, NOx and VOC emissions from the OTR region and primary $PM_{2.5}$ emissions from the Philadelphia MSA would need to be significantly reduced (Table 3). The results of the OPERA calculation also show that reductions in SO_2 emissions were not cost-effective for achieving the ozone and $PM_{2.5}$ air quality target; this was because reductions in SO_2 emissions would only decrease $PM_{2.5}$ levels but reductions in NOx and VOC could decrease both ozone and $PM_{2.5}$ concentrations in Philadelphia. Overall, a 42% reduction in controllable NOx emissions and a 38% reduction in controllable VOC emissions from the OTR region, as well as a 34% reduction in controllable primary $PM_{2.5}$ emissions from the Philadelphia MSA, would be the most cost-effective control strategy for achieving the prescribed ozone and $PM_{2.5}$ air quality targets on August 2, 2007, in Philadelphia (Table 3). The least cost of the emission

Table 2. Daily-average $PM_{2.5}$ concentrations ($\mu g/m^3$) as well as first, second, and cross sensitivities to SO_2, NOx, and VOC and primary $PM_{2.5}$ emissions on August 2, 2007

Region	Sensitivity/$PM_{2.5}$	Atlanta 16.10	Chicago 26.25	D.C. 11.73	New York 38.83	Philadelphia 30.76
OTR	1st NOx	0.23	0.05	1.54	5.00	8.81
	1st VOC	–0.02	–0.03	–0.22	0.17	–0.48
	2nd NOx	–0.14	–0.02	–0.50	–1.29	–1.56
	2nd VOC	0.01	~0	0.04	~0	0.06
	2nd NOx_VOC	~0	~0	–0.03	0.04	–0.02
	1st SO_2	0.67	0.12	1.65	1.25	2.82
LADCO	1st NOx	~0	2.31	0.02	0.06	0.24
	1st VOC	~0	–0.40	–0.01	–0.01	–0.06
	2nd NOx	0.01	–1.63	–0.01	–0.01	–0.04
	2nd VOC	~0	0.04	~0	~0	~ 0
	2nd NOx_VOC	~0	0.03	~0	~0	~ 0
	1st SO_2	0.16	2.96	0.07	0.04	0.12
CENRAP	1st NOx	~0	0.25	0.01	0.02	0.11
	1st VOC	~0	–0.02	~0	~0	–0.01
	2nd NOx	~0	–0.03	~0	~0	–0.02
	2nd VOC	~0	~0	~0	~0	~ 0
	2nd NOx_VOC	~0	~0	~0	~0	~ 0
	1st SO_2	0.04	0.27	0.01	0.01	0.02
SEMAP	1st NOx	0.63	0.01	~0	~0	~ 0
	1st VOC	–0.01	~0	~0	~0	~ 0
	2nd NOx	0.11	~0	~0	~0	~ 0
	2nd VOC	~0	~0	~0	~0	~ 0
	2nd NOx_VOC	–0.01	~0	~0	~0	~ 0
	1st SO_2	0.77	0.03	0.03	~0	0.01
Primary $PM_{2.5}$		3.83	9.1	3.57	10.52	7.08

reductions for achieving the air quality target in Philadelphia was about $5.6 billion (1999 dollars). In the case study, Philadelphia and New York are the two cities that need to achieve the NAAQS for both O_3 and $PM_{2.5}$. OPERA is able to identify the least-cost control strategies for Philadelphia but not New York. This is because the O_3 and $PM_{2.5}$ levels in New York were much higher than those in Philadelphia, and the required magnitudes of emission controls for New York were out of feasible ranges of the emission reductions.

Least-cost ozone and $PM_{2.5}$ air quality management strategies for multiple cities

For real-world applications, air quality managers may set multiple targets of reductions in air pollutant levels according to different air quality and human health objectives. To show the applicability of OPERA for developing optimal control measures for multiple locations, least-cost emission control strategies for simultaneously achieving the 75 ppb MDA8h ozone and 25 $\mu g/m^3$ daily $PM_{2.5}$ targets in Atlanta, Chicago, Washington, DC, and Philadelphia are examined in this section. New York is not considered in this analysis since it was infeasible to achieve the prescribed air quality targets due to high background ozone and/or significant amount of transported $PM_{2.5}$ (discussed in the previous section). To identify the least-cost control strategies for achieving the ozone and $PM_{2.5}$ air quality targets in the four cities simultaneously, the required reductions in ozone and $PM_{2.5}$ concentrations (i.e., baseline concentrations – target concentrations) for all the four cities should be included in the constraints of OPERA (eqs 2 and 3). In this case study, 24 constraints are used in the OPERA calculations because eight equations are used to define ozone and $PM_{2.5}$ reduction targets for the four cities, 12 equations are used to constrain reduction ratios of emissions from the four regions, and four equations are applied to limit reduction ratios of primary $PM_{2.5}$ emitted from the four MSAs.

To achieve the ozone and $PM_{2.5}$ air quality targets for the four cities simultaneously, NOx and VOC emissions from OTR, NOx from SEMAP, and primary $PM_{2.5}$ emissions from the Chicago and Philadelphia MSAs would need to be significantly reduced (Table 3). The results of the OPERA calculation show that NOx and VOC emissions from OTR would need to be reduced by about 42% and 37%, respectively, to achieve the prescribed air quality targets in the four cities simultaneously. In addition to the reduction in the emissions from OTR, a 23% reduction in controllable NOx emissions from SEMAP would

Table 3. Least-cost control strategies for achieving 75 ppb MDA8h ozone and 25 μg/m^3 daily average $PM_{2.5}$ targets[1] on August 2, 2007, for single cities and multiple cities (Atlanta, Chicago, Washington, DC, and Philadelphia) as a whole, as percent relative to total controllable emissions

	Atlanta	Chicago	Washington, DC	New York	Philadelphia	Four cities[2]
Reduction (%) in controllable SO_2 emissions						
OTR	~0	~0	~0	infeasible	~0	~0
LADCO	~0	~0	~0	infeasible	~0	~0
CENRAP	~0	~0	~0	Infeasible	~0	~0
SEMAP	~0	~0	~0	Infeasible	~0	~0
Reduction (%) in controllable NOx emissions						
OTR	15.8	~0	20.7	Infeasible	41.6	41.9
LADCO	1.6	0.3	0.8	infeasible	7.9	8.0
CENRAP	0.8	~0	0.5	Infeasible	5.3	5.4
SEMAP	38.9	~0	~0	Infeasible	~0	22.9
Reduction (%) in controllable VOC emissions						
OTR	0.5	~0	~0	Infeasible	37.5	37.2
LADCO	~0	~0	~0	Infeasible	~0	~0
CENRAP	~0	~0	~0	Infeasible	~0	~0
SEMAP	3.4	~0	~0	infeasible	~0	0.7
Reduction (%) in primary $PM_{2.5}$ emissions						
Atlanta	~0	~0	~0	Infeasible	~0	~0
Chicago	~0	13.9	~0	Infeasible	~0	11.5
D.C.	~0	~0	~0	Infeasible	~0	~0
New York	~0	~0	~0	infeasible	~0	~0
Philadelphia	~0	~0	~0	Infeasible	34.2	33.8
Cost (millions of 1999 dollars)	1,333	253	838	—	5,639	5831

[1]If baseline MDA8h ozone and/or daily average $PM_{2.5}$ concentrations were below the air quality targets, zero reductions are used in OPERA calculations.
[2]The target was to achieve 75 ppb MDA8h ozone and 25 μg/m^3 daily average $PM_{2.5}$ in Atlanta, Chicago, Washington, DC, and Philadelphia simultaneously.

be needed to decrease the baseline MDA8h ozone concentrations to 75 ppb in the four cities. Reductions in primary $PM_{2.5}$ emissions from the Chicago (~12%) and Philadelphia (~34%) MSAs are also found to be cost-effective for attaining prescribed reductions in $PM_{2.5}$ levels in the four cities. On the other hand, the results show that reductions in SO_2 emissions were less cost-effective than NOx, VOC, and primary $PM_{2.5}$ emission controls for achieving the ozone and $PM_{2.5}$ air quality targets in the four cities. This was because SO_2 emission reductions would only decrease $PM_{2.5}$ concentrations and reductions in primary $PM_{2.5}$ emissions were more cost-effective than SO_2 emission controls. The least cost for achieving the ozone and $PM_{2.5}$ air quality targets in the four cities was about $5.8 billion (1999 dollars); such high costs were mainly because of significant reductions in regional NOx and VOC as well as local primary $PM_{2.5}$ emissions (Table 3). Overall, the result shows that the cost of considering multiple cities simultaneously was much less than the cost of adding up across the individual city estimates (~$8 billion). This is because controls of precursor emissions targeted to decrease air pollutant levels in a city could also reduce air pollutant levels in other cities. For example, controls of NOx emissions from the OTR region targeted to reduce air pollutant levels in Philadelphia could also reduce air pollutant concentrations in the other four cities examined in the case study.

Limitations and uncertainties

The results of OPERA could be significantly affected by the cost function and sensitivities used in the calculation. As discussed earlier, AirControlNET is a somewhat dated tool that may not fully reflect currently available controls. A state-of-art cost analysis tool should be used to estimate the cost function when it is available in the future. CMAQ-HDDM is used in this study, but other models and approaches (e.g., CAMx-HDDM, CMAQ-Adjoint, CMAQ with the brute force method, etc.) can also be used to estimate responses of air pollutants to changes in precursor emissions. Careful investigations of costs of emission control measures and responses of air quality to emission reductions would be extremely important when using OPERA in designing multipollutant air quality management strategies for different regions.

It is also important to note that the results of the case study may not be robust or meaningful since only one day of air quality was investigated. Longer terms of air quality (e.g., June–August for 3 consecutive years) should be investigated when OPERA is used in developing air pollution mitigation strategies. The ignorance of co-benefits of multiple-precursor controls could cause biases in the results. Furthermore, biases in the modeled ozone and particulate matter concentrations (e.g., underestimation of SOAs) and sensitivities could also

affect the results of the case study. Because we only included a limited set of cross-pollutant sensitivities, specifically between NOx and VOC, and did not include cross-pollutant sensitivities between PM precursors, specifically SO_2 and NOx, our conclusions about the optimality of applying direct PM controls instead of SO_2 controls are subject to potentially significant uncertainties, especially when combined with uncertainties in the relative costs of controls. The potential biases are not specifically examined here since the case study is to demonstrate the proposed optimization model (i.e., OPERA).

Conclusions

We developed an optimization-based model, OPtimal integrated Emission Reduction Alternatives (OPERA), which allows identification of least-cost control strategies for achieving multipollutant air quality targets at multiple locations simultaneously. Two main pieces of input are needed for OPERA: (1) sensitivities of air quality to emission changes and (2) cost functions of emission reductions. To demonstrate how to develop air quality management strategies using OPERA, we presented a case study focusing on ozone and $PM_{2.5}$ air quality in the summer of 2007 in five major cities in the eastern United States. From the results of the case study, we have the following central findings in this study. First, emission reductions from distant regions could be cost-effective for achieving prescribed ozone and $PM_{2.5}$ levels in the cities examined. Second, reducing regional NOx and VOC as well as local primary $PM_{2.5}$ emissions was more cost-effective than controlling SO_2 emissions for decreasing ozone and $PM_{2.5}$ concentrations simultaneously. This was because reductions in SO_2 emissions would only decrease $PM_{2.5}$ concentrations, and reductions in primary $PM_{2.5}$ emissions were more cost-effective than SO_2 emission controls. A major strength of OPERA is its flexibility, which allows for changes in regulations, involving agencies, study regions, and so on to be readily incorporated. Overall, OPERA has been demonstrated to be efficient to develop least-cost emission control strategies for achieving multipollutant air quality targets at multiple locations simultaneously and could be useful for policymakers developing integrated air quality management plans.

For developing "optimal" air quality management strategies, another approach is to maximize environmental and/or human health benefits when specific constraints (e.g., budgets) are considered in decision-making processes. Such an approach is being developed by the authors, and a benefit maximization model will be available in the near future.

Supplemental Material

Supplemental data for this article can be accessed at http://dx.doi.org/10.1080/10962247.2015.1014073.

Acknowledgments

The authors acknowledge the Texas Advanced Computing Center (TACC) at the University of Texas at Austin for providing high-performance computing (HPC) resources that have contributed to the research results reported within this study.

Funding

The authors thank the EPA for providing funding under the National Center for Environmental Research (NCER) STAR Program grant R835218. Although the research described in the paper has been funded by the EPA STAR program, it has not been subjected to any EPA review and therefore does not necessarily reflect the views of the agency, and no official endorsement should be inferred.

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About the Authors

Kuo-Jen Liao is an assistant professor of environmental engineering at Texas A&M University–Kingsville in Kingsville, Texas.

Xiangting Hou is a Ph.D. candidate in the Department of Environmental Engineering at Texas A&M University–Kingsville in Kingsville, Texas.