Journal of Exposure Analysis and Environmental Epidemiology (2005) 15, 439–457
© 2005 Nature Publishing Group All rights reserved 1053-4245/05/$30.00
www.nature.com/jea

A source-to-dose assessment of population exposures to fine PM and ozone in Philadelphia, PA, during a summer 1999 episode

PANOS G. GEORGOPOULOS,[a] SHENG-WEI WANG,[a] VIKRAM M. VYAS,[a] QING SUN,[a] JANET BURKE,[b] RAM VEDANTHAM,[b] TOM McCURDY[b] AND HALÜK ÖZKAYNAK[b]

[a]*Environmental and Occupational Health Sciences Institute, Piscataway, New Jersey, USA*
[b]*USEPA-National Exposure Research Laboratory, Research Triangle Park, North Carolina, USA*

A novel source-to-dose modeling study of population exposures to fine particulate matter ($PM_{2.5}$) and ozone (O_3) was conducted for urban Philadelphia. The study focused on a 2-week episode, 11–24 July 1999, and employed the new integrated and mechanistically consistent source-to-dose modeling framework of MENTOR/SHEDS (Modeling Environment for Total Risk studies/Stochastic Human Exposure and Dose Simulation). The MENTOR/SHEDS application presented here consists of four components involved in estimating population exposure/dose: (1) calculation of ambient outdoor concentrations using emission-based photochemical modeling, (2) spatiotemporal interpolation for developing census-tract level outdoor concentration fields, (3) calculation of microenvironmental concentrations that match activity patterns of the individuals in the population of each census tract in the study area, and (4) population-based dosimetry modeling. It was found that the 50th percentiles of calculated microenvironmental concentrations of $PM_{2.5}$ and O_3 were significantly correlated with census-tract level outdoor concentrations, respectively. However, while the 95th percentiles of O_3 microenvironmental concentrations were strongly correlated with outdoor concentrations, this was not the case for $PM_{2.5}$. By further examining the modeled estimates of the 24-h aggregated $PM_{2.5}$ and O_3 doses, it was found that indoor $PM_{2.5}$ sources dominated the contributions to the total $PM_{2.5}$ doses for the upper 5 percentiles, Environmental Tobacco Smoking (ETS) being the most significant source while O_3 doses due to time spent outdoors dominated the contributions to the total O_3 doses for the upper 5 percentiles. The MENTOR/SHEDS system presented in this study is capable of estimating intake dose based on activity level and inhalation rate, thus completing the source-to-dose modeling sequence. The MENTOR/SHEDS system also utilizes a consistent basis of source characterization, exposure factors, and human activity patterns in conducting population exposure assessment of multiple co-occurring air pollutants, and this constitutes a primary distinction from previous studies of population exposure assessment, where different exposure factors and activity patterns would be used for different pollutants. Future work will focus on incorporating the effects of commuting patterns on population exposure/dose assessments as well as on extending the MENTOR/SHEDS applications to seasonal/annual studies and to other areas in the U.S.
Journal of Exposure Analysis and Environmental Epidemiology (2005) **15,** 439–457. doi:10.1038/sj.jea.7500422; published online 16 February 2005

Keywords: *ozone, PM, exposure, dose, modeling, MENTOR, SHEDS.*

Introduction

Evaluation of human exposure to complex atmospheric contaminants such as ozone (O_3), fine airborne particulate matter ($PM_{2.5}$), and primary and secondary air toxics, requires characterization of the concentrations and physico-chemical attributes of these contaminants (a) at the local (e.g. census tract or "neighborhood") scale, (b) in residential and occupational environments, and (c) in the air flow that actually enters the human respiratory tract (personal air). Models for estimating population exposures to atmospheric pollutants have been developed by USEPA for regulatory purposes. In fact, USEPA offices (OAQPS and NERL) have supported comprehensive efforts in this area, resulting in the development and various applications of the NEM/pNEM/APEX3 (NAAQS Exposure Model, Probabilistic NAAQS Exposure Model, and Air Pollution Exposure Model) (Johnson et al., 1996; Glen, 2002), HAPEM (Hazardous Air Pollutant Exposure Model) (Rosenbaum, 2002) and SHEDS (Stochastic Human Exposure and Dose Simulation) (Burke et al., 2001) families of models.

The NEM/pNEM/APEX3 and SHEDS families of models typically provide hourly averages of exposure estimates, defined by concentration and breathing rate for each individual exposure event; these estimates are then aggregated to other time periods of interest (daily, weekly, annually, etc.). pNEM and SHEDS implementations also simulate certain aspects of the variability and uncertainty in the principal factors affecting exposure. An alternative approach is taken by the HAPEM family of models that typically provide long term (e.g., annual) average exposure estimates, based on the quantity of time spent per year in each

1. Address all correspondence to: Panos G. Georgopoulos, 170 Frelinghuysen Road, Piscataway, NJ 08854, USA. Tel.: +1 732 445 0159; Fax: +1 732 445 0915. E-mail: panosg@fidelio.rutgers.edu
Received 12 May 2003; accepted 17 December 2004; published online 16 February 2005

combination of geographic locations and microenvironments (Rosenbaum, 2002). The various NEM- and SHEDS-type models are therefore expected to be more appropriate for pollutants with nonlinear dynamics, such as ozone and aerosol, and provide approaches for enhancing related health assessments, while the general HAPEM approach offers advantages in combining longer-term assessments.

Inputs for these models include:

- ambient pollutant concentrations from monitoring data or from air quality model output,
- data on indoor/outdoor relationships for ambient pollutants,
- demographic data for the area and/or population of interest,
- human activity pattern data,
- age- and gender-specific data for estimating inhalation rates based on physical activity level.

A mechanistic framework for linking available models and databases would improve inhalation exposure assessment in terms of:

- providing consistency,
- allowing assessments on multiple scales, and incorporating the processes occurring from the emission source to the dose received by individual exposures,
- optimizing the usage of the most up-to-date models and databases.

The Modeling Environment for Total Risk Studies (MENTOR) system has been configured for the needs of this study so as to provide an integrated and mechanistically consistent source-to-dose modeling framework for assessment of simultaneous exposures to fine PM and O_3. The objective of the overall MENTOR project has been to develop, apply and evaluate state-of-the-art methods and computational tools for a wide range of environmental applications, that utilize existing models when available or provide new approaches to "fill gaps" in the source-to-dose sequence. So, MENTOR links state-of-the-art predictive models of exposure and dose, coupled with up-to-date national, regional, and local databases of environmental, microenvironmental, biological, physiological, demographic, etc. parameters. MENTOR is not a "new model"; it is an evolving open computational toolbox intended to facilitate consistent multiscale source-to-dose modeling of exposures to (atmospheric or multimedia) contaminants, for individuals and populations.

Two implementations of the MENTOR system are currently available: MENTOR/SHEDS-1A (MENTOR using the Stochastic Human Exposure and Dose Simulation (SHEDS) approach in a "One Atmosphere (1A) setting) characterizes simultaneous exposures to multiple atmospheric contaminants taking into account their physical and chemical interactions for individuals and/or populations (Georgopoulos et al., 2004a; USEPA, 2004a). MENTOR/SHEDS-4M (MENTOR using the Stochastic Human Exposure and Dose Simulation (SHEDS) approach for Multiple co-occurring contaminants and Multimedia, Multipathway, Multiroute exposures (4M)) quantifies aggregate and cumulative exposures and doses of individuals and populations to multiple contaminants (Georgopoulos et al., 2004b; USEPA, 2004b).

Models and databases currently linked with the MENTOR/SHEDS-1A system, as configured for the present study, include:

- *Emissions processing*: Sparse Matrix Kernel Estimator (SMOKE) (Houyoux and Vukovich, 1999) and Emissions Modeling System for Hazardous Air Pollutants (EMS-HAP) (Strum et al., 2002),
- *Meteorological models*: Mesoscale Model 5 (MM5) (Grell et al., 1994; NCAR, 1999), Regional Atmospheric Modeling System (RAMS) (Walko and Tremback, 2001), and California Meteorological Model (CALMET) (Scire et al., 2000),
- *Regional-scale and local-scale air quality modeling systems that include*: EPA's Models-3/Community Multi-scale Air Quality (CMAQ) (USEPA, 1999b), Multiscale Air Quality Simulation Platform (MAQSIP) (Odman and Ingram, 1996), Urban Airshed Model (UAM-V) (Douglas et al., 2000), Comprehensive Air Quality Model With Extensions (CAMx) (ENVIRON, 2002), and PM-CAMx update (Morris et al., 2002),
- *Advanced optimal interpolation methods based on*: Spatio-temporal Random Field (STRF) and Bayesian Maximum Entropy (BME) (Serre and Christakos, 1999) theories,
- *Exposure models*: Stochastic Human Exposure and Dose (SHEDS), pNEM/APEX, HAPEM,
- Inhalation dosimetry models,
- *Databases*: Consolidated Human Activity Database (CHAD) (McCurdy et al., 2000), 1990 US Census data (US Census Bureau, 1992), National Emissions Inventory (NEI) 1999 (USEPA, 2001b), National Emission Trends (NET) 1998 (USEPA, 2000), Aerometric Information Retrieval System (AIRS) (USEPA, 2001a), and National Climatic Data Center (NCDC) (NOAA, 2003).

The MENTOR/SHEDS-1A system was applied in this study to model population exposures to fine particulate matter ($PM_{2.5}$) and O_3 for urban Philadelphia (see Figure 1b). The study focused on a 2-week period, 11–24 July 1999, and employed USEPA's Models-3/CMAQ with MM5 (NCAR, 1999; USEPA, 1999b) and SMOKE (Houyoux and Vukovich, 1999), STRF/BME modules, CHAD, a generalized version of SHEDS recoded for incorporation into MENTOR (MENTOR/SHEDS), and a newly developed dosimetry model, in order to achieve consistent source-to-dose analysis. The air pollution episode considered in this study was captured in both the newly expanded regional air-quality monitoring networks (AIRS, etc.) (USEPA, 2002) and the field measurements of the

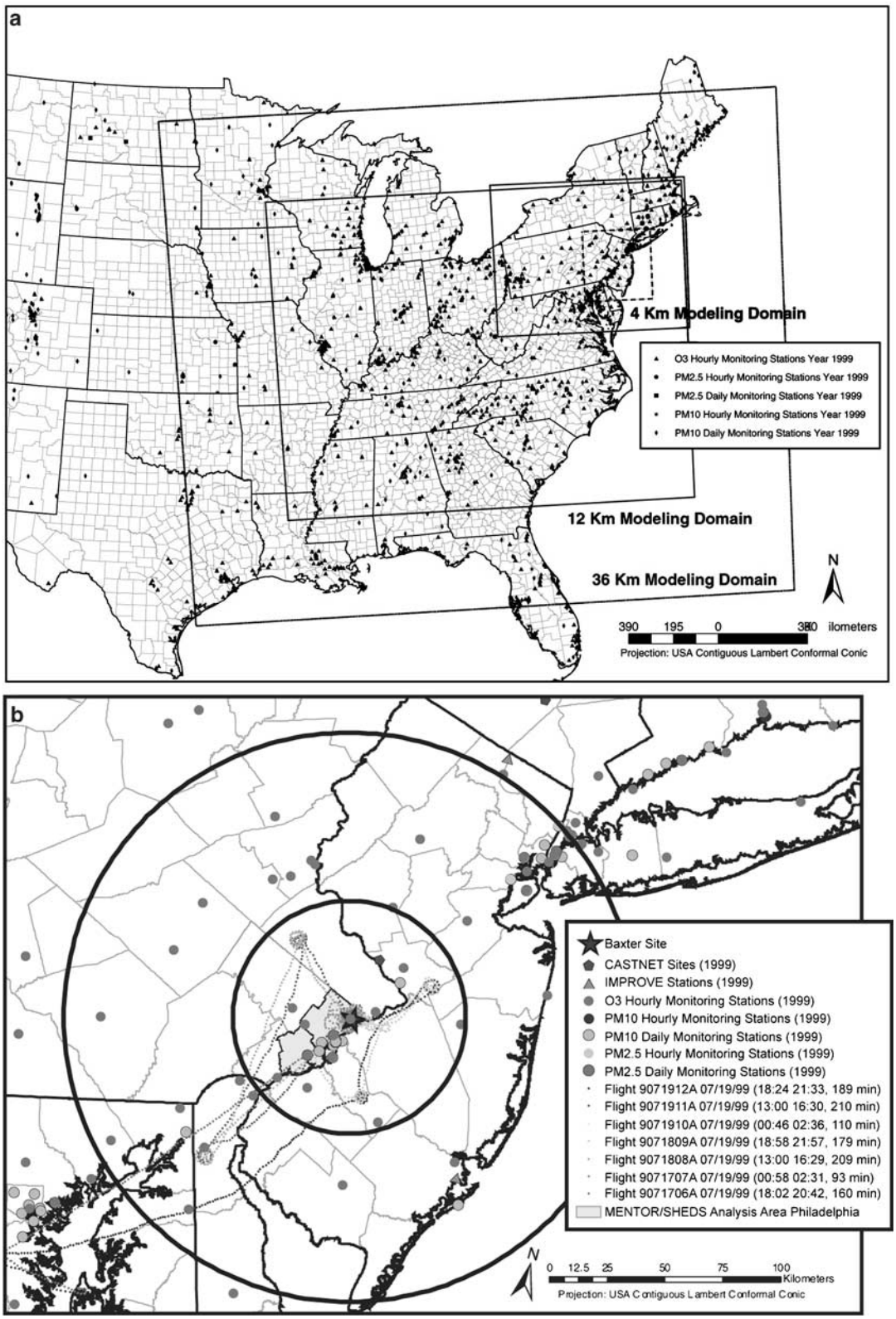

Figure 1. (a) Map of Models-3/CMAQ air quality modeling domain with 36, 12, and 4-km horizontal grid resolutions employed in the current applications. (b) Air quality monitoring stations and tracks of air quality monitoring flights undertaken as part of the NE-OPS study, in the vicinity of this case study: area in gray (urban Philadelphia) represents focus of the MENTOR/SHEDS analysis of population exposures to O_3 and fine PM during the summer of 1999. The circles in the figure correspond to radii of 50 and 100 km with the Baxter NE-OPS site at the center.

North East Oxidant and Particulate Study (NE-OPS) (Philbrick, 1999) of the summer of 1999. This extensive database of observational information has provided the opportunity to evaluate individually various components of the comprehensive modeling system that was used to assess population exposures to $PM_{2.5}$ and O_3 and thus build confidence regarding the applicability of the methods used. Furthermore, it should be noted that past studies have often been limited in their scope because they typically addressed only one parameter (O_3 or $PM_{2.5}$) (Hayes, 1989, 1991; Lurmann et al., 1992; Lurmann and Korc, 1994; Johnson et al., 1996). The current study attempts, however, an assessment of simultaneous population exposures due to co-occurring O_3 and $PM_{2.5}$.

Methods

The comprehensive modeling of individual/population exposure to ozone and aerosol in general requires the seven steps (or components, as some of them do not have to be performed in sequence), that are listed below. This list of steps represents a "composite" based on frameworks described in the literature over the last 20 years (Ott, 1982, 1985; Lioy, 1990; USEPA, 1992; Georgopoulos and Lioy, 1994; McCurdy, 1994; USEPA, 1997) as well as on the structure of various existing inhalation exposure models (NEM/pNEM, HAPEM, SHEDS, REHEX, EDMAS, MENTOR, APEX, AIRPEX, AIRQUIS, etc. (Whitfield et al., 1997; Freijer et al., 1998; Clench-Aas et al., 1999; USEPA, 1999a; Georgopoulos and Lioy, 2000; Rifai et al., 2000; Burke et al., 2001; Rosenbaum, 2002)) that have been used in past or in on-going studies:

(1) Estimation of background levels of air pollutants through either (or a combination of):
   (a) multivariate spatiotemporal analysis of monitor data,
   (b) emissions-based air quality modeling (with regional, grid-based models such as Models-3/CMAQ, CAMx and REMSAD).
(2) Estimation of local outdoor pollutant levels that characterize locally the ambient air of an administrative unit (such as a census tract) or a conveniently defined grid through either
   (a) spatiotemporal statistical analysis of monitor data,
   (b) subgrid "corrections" of multiscale model estimates,
   (c) application of a local scale air quality model such as ISCST, AERMOD, etc.
(3) Characterization of attributes of populations (geographic density, age, gender, race, income, etc.)
   (a) select fixed-size sample population that statistically reproduces essential demographics or
   (b) divide population of interest into exhaustive set of cohorts.
(4) Development of activity event (or exposure event) sequences for each member of the sample population or for each cohort for the exposure period through either:
   (a) existing databases from composites of past studies (for baseline assessment),
   (b) study-specific information (special registries).
(5) Estimation of levels and temporal profiles of pollutants in various microenvironments (streets, residences, offices, restaurants, vehicles, etc.) through one (or a combination) of the following methods:
   (a) regression of observational data,
   (b) simple linear mass balance,
   (c) detailed (nonlinear) gas/aerosol chemistry models,
   (d) detailed combined chemistry and fluid dynamics models.
(6) Calculation of appropriate inhalation rates for the members of the sample population combining the physiological attributes of the study subjects and the activities pursued during the individual exposure events.
(7) Calculation of target tissue dose through biologically based modeling (e.g., physiologically based respiratory deposition modeling for PM or toxicokinetic modeling for organic air toxics) when the structure of the modeling system allows it.

A generalized version of SHEDS (MENTOR/SHEDS-1A) was developed in this study to estimate exposures and doses of $PM_{2.5}$ and O_3 for the population of Philadelphia during an air quality episode. MENTOR/SHEDS-1A keeps the following stochastic features as in previous version of SHEDS (Burke et al., 2001):

- generation of simulation population based on census demographics,
- development of exposure event sequences for each individual in the simulated population by selecting from diaries that match the demographic characteristics of the simulated individual, and
- calculation of $PM_{2.5}$ microenvironmental concentration for each location in the activity diary using microenvironment-specific equations and randomly selected values from distributions for the various parameters of the equations.

The following additional features of MENTOR/SHEDS-1A were developed and used in this study to characterize exposures and doses to co-occurring O_3 and $PM_{2.5}$:

- calculation of microenvironmental O_3 concentrations for each location in the activity diary, using microenvironment-specific equations and randomly selected values from distributions of the various parameters of the equations,
- estimation of inhalation rates for each exposure event in the sequence, based on physical activity level, and calculation of exposure and dose for each exposure event, based on microenvironmental concentration, time spent in microenvironment, and the inhalation rate

corresponding to the activities of the individual in this microenvironment.

The following describe how the above steps of comprehensive population exposure modeling have been implemented within the MENTOR/SHEDS-1A framework in the current study. An overview flowchart of the structure of the MENTOR/SHEDS-1A approach as implemented in the present study is shown in Figure 2.

Step 1: The Eulerian Community Multi-scale Air Quality (CMAQ) model (USEPA, 1999b) was used to simulate ambient concentrations for the time period of interest. The 1998 version of the National Emission Trends (NET) database maintained by USEPA was used for the area, mobile and point source terms (USEPA, 2001b), as the 1999 (final draft) release of the National Emissions Inventory (NEI) was not available yet. The emissions were processed using the Sparse Matrix Kernel Estimator (SMOKE) preprocessor (Houyoux and Vukovich, 1999). Meteorological inputs were obtained from the National Climatic Data Center (NCDC) archives, and incorporated ("assimilated") into the calculations of the National Center for Atmospheric Research (NCAR) MM5 Version 3 mesoscale meteorological model (NCAR, 1999). The MM5 model utilized as input gridded European Centre for Medium-Range Weather Forecasts (ECMWF) data at 2.5° resolution as well as rawinsonde and surface observations. The model utilized the high-resolution Blackadar scheme for Planetary Boundary Layer (PBL), Grell's scheme for cumulus parameterization, mixed phase (Reisner scheme) for explicit moisture and a cloud radiation scheme. The output frequency for the MM5 and CMAQ simulations was 1 h. The CMAQ simulations were carried out for the 11 July 1999 00 Coordinated Universal Time (UTC) to 25 July 1999 12 UTC period. Three levels of nested grids were used with grid resolutions of 36, 12 and 4 km. The 4-km domain encompassed the Philadelphia and New Jersey regions while the 36-km domain encompassed the entire Ozone Transport Assessment Group (OTAG) (Guinnup and Collom, 1997) domain (Eastern United States) as shown in Figure 1a. The grid dimensions in the east–west and north–south directions are 69×57, 46×46 and 61×70 at the 36, 12 and 4-km grid resolutions, respectively. In the vertical direction, a nonhydrostatic coordinate was used with 14 layers centered at the following values of the sigma-p coordinate: 0.9975, 0.9925, 0.985, 0.9725, 0.955, 0.9325, 0.9, 0.84, 0.75, 0.65, 0.525, 0.375, 0.225 and 0.075. The chemical mechanism employed was RADM2 (Stockwell et al., 1990, 1997) with modal aerosol dynamics and aqueous chemistry, which included 59 gas phase species, 26 aerosol species and 161 reactions. Details of the regional photochemical modeling study are available in Chandrasekar

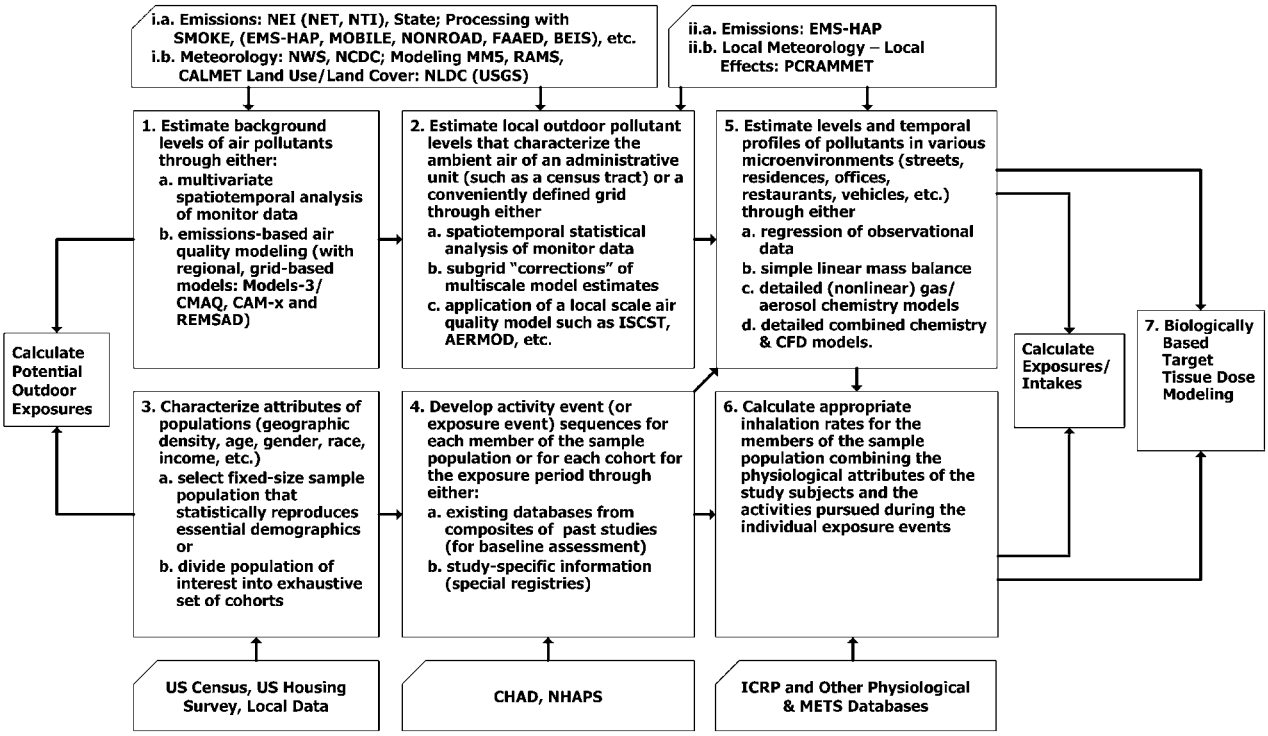

Figure 2. A generalized 7-step flowchart describing the processes involved in assessing population exposures/doses in a source-to-dose framework; this is also referred to as person-oriented population-based exposure modeling (POM/PBEM). This flowchart reflects the structure of the MENTOR/SHEDS approach, but also provides a general "template" for comparing the application of exposure assessment systems. A subset of the models appearing in this flowchart has been used in the present study.

et al. (2001a, b), Sun and Georgopoulos (2002), and Sun et al. (2002).

Air quality monitor data were obtained for the region and time period of interest from the Aerometric Information Retrieval System (AIRS) maintained by USEPA. Additional measurements were also obtained from the field intensive study conducted by the NE-OPS consortium (Philbrick et al., 2002). Figure 1a depicts the geographical locations of both O_3 and PM monitoring stations in the regional air quality modeling domain of the present study. For performing geostatistical analysis of monitor data, observations were available from 32 ozone hourly monitoring stations (including one NE-OPS station) operating within a radius of 100 km from urban Philadelphia; and 4 $PM_{2.5}$ (1 h averaged) monitoring stations (including the NE-OPS station) within the same region. Locations of the monitors that were utilized are shown in Figure 1b. The observational data from the O_3 monitoring stations were used in conjunction with novel spatiotemporal interpolation approaches in this study.

Step 2: The regional air quality modeling predictions and the observed data were interpolated to the scale of census tracts by using optimal spatiotemporal methods, to link regional scale predictions to local (census tract) scale outdoor concentrations, in order to be used as inputs for the population exposure calculations. A particular issue considered in this study, is that of the proper interpolation of monitor or model predicted concentrations to the finer resolution of census tract or neighborhood. It is well known that the method of interpolation may influence the estimates of population exposures by acting as a filter on extreme events; here the application of both Spatiotemporal Random Field (STRF) theory and of Bayesian Maximum Entropy (BME) methods seeks to address this issue by providing alternative approaches that do not act as filters to concentrations at the tails of the spatiotemporal distribution or that could even capture "subgrid" effects through appropriate utilization of combined spatial and temporal information.

This STRF estimation method was conceptualized by Christakos (1991), and developed by Vyas and Christakos (1997). A spatiotemporal random field of order v in space and μ in time (STRF-v/μ) is a spatially nonhomogeneous and temporally nonstationary random field $X(\mathbf{s}, t)$ for which a mathematical operation Q can be defined in space/time that transforms $X(\mathbf{s}, t)$ to a zero-mean space homogeneous/time stationary process $Y(\mathbf{s}, t) = Q[X(\mathbf{s}, t)]$. By its construction, the Q operation filters out spatiotemporal trends such as linear or polynomial trends in space, time or unified space/time. Other forms of trends including exponential or trigonometric functions can also be considered. The orders v and μ are called space/time continuity orders because they characterize the rate of change of the functions modeling trends in the distribution of the measurements on the environmental process being studied.

These theoretical constructs $Y(\mathbf{s}, t)$ and $X(\mathbf{s}, t)$ are then used to formulate a spatiotemporal kriging method. The estimate $\hat{X}(\mathbf{s}, t)$ of the environmental process $X(\mathbf{s}, t)$ at point $\mathbf{s}$ and instant t for which measurements are not available is given by

$$\hat{X}(s_k, t_k) = \Xi^{\mathrm{T}} X$$

where Ξ is a vector of weights associated with the data points such that the error variance is given by the expression

$$\sigma_x^2 = \overline{\left[\hat{X}(s_k, t_k) - X(s_k, t_k)\right]^2}$$

These kriging weights are obtained by solving the system of equations

$$K\Xi^* = \Theta$$

where K is a matrix of the generalized covariances between all data points/instants and the corresponding space/time trends; Θ is a vector that includes covariances between the data and the points/instants under estimation and also the space/time trends at the latter points/instants; Ξ^* is the vector of weights Ξ and Lagrange multipliers introduced so that the constraints of STRF-v/μ are satisfied. In addition to the STRF method, the BME method (Serre and Christakos, 1999) was also used in this study to obtain census tract scale interpolations of CMAQ predictions. Details of the comparative application of the STRF and BME methods are available in Georgopoulos et al. (2004a).

Step 3: The attributes of the population under study were retrieved from the 1990 U.S. Census Survey. Due to the variability of the urban population a rather large statistical sample of 500 "virtual individuals" was sampled for each of the 482 census tracts under study to statistically reproduce essential demographics distributions of age, gender, housing type and employment status.

Step 4: A 24-h activity diary for each "virtual individual" of the simulated population was selected from the CHAD diaries so as to match the demographic characteristics of the virtual individual with respect to age, gender, employment, and smoking status. An activity diary is a sequence of events that simulate the movement of a "virtual individual" through geographic locations and microenvironments during the simulation period. Each event is defined by geographic location, start time, duration, microenvironment visited, and an activity performed. There are 113 microenvironments in the CHAD diaries. These microenvironments are grouped into nine categories in the current study: home, other indoor, outdoor, vehicle, school, office, store, restaurant, and bar.

Step 5: The outdoor concentrations of O_3 and $PM_{2.5}$, interpolated at census-tract level, are used as inputs to the MENTOR/SHEDS-1A modules for estimating different microenvironmental concentrations. The estimation of O_3

concentrations in various microenvironments is based on the simple mass balance equation used in pNEM/O_3 model approach as below:

$$\frac{dC_{in}}{dt} = (F_p)(\text{ach})(C_{out}) + \frac{S}{V} - (\text{ach} + F_d)(C_{in}) \quad (1)$$

where C_{in} is the indoor concentration (mass/volume), F_p the penetration factor (dimensionless fraction), ach the air exchange rate (1/time), C_{out} the outdoor concentration (mass/volume), S the indoor source generation rate (mass/time), V the indoor volume (volume), and F_d the O_3 decay rate (1/time). This equation is further simplified on the basis of the following assumptions: (1) steady-state, (2) $S = 0$ (no indoor sources), and (3) $F_p = 1$. Then one has

$$\frac{C_{in}}{C_{out}} = \frac{\text{ach}}{\text{ach} + F_d} \quad (2)$$

For different microenvironments, different distributions for determining the parameters of ach and F_d are used. The values of these two parameters (ach and F_d) are drawn randomly from their corresponding distributions according to the locations of activity events.

PM concentrations in the indoor and in-vehicle microenvironments are calculated using microenvironment-specific equations for the relationship between outdoor and indoor PM concentrations. For the indoor residential microenvironment, a single-compartment, steady-state mass balance equation (Özkaynak et al., 1996) is used to calculate indoor PM concentrations from the combination of infiltration of ambient PM indoors and indoor PM sources:

$$C_{residential} = \frac{P \times \text{ach}}{\text{ach} + k} C_{ambient} + \frac{E_{smk} N_{cig} + E_{cook} t_{cook} + E_{other} t_s}{(\text{ach} + k) V t_s} \quad (3)$$

where $C_{ambient}$ is the ambient outdoor PM concentration (μg/m^3), P the penetration factor (unitless), k the deposition rate (h^{-1}), ach the air exchange rate (h^{-1}), E_{smk} the emission rate for smoking (μg/cig), N_{cig} the number of cigarettes smoked during model time step, E_{cook} the emission rate for cooking (μg/min), t_{cook} the time spent cooking during model time step (min), E_{other} the emission rate for other source (μg/h), t_s the model time step (h), and V the residential volume (m^3). The first term in Equation (3) describes the infiltration of ambient PM indoors. The second term describes the generation of particles from indoor sources. These parameters mentioned above are randomly drawn from their specific distributions except the variable t_s, which is based on the duration of the activity events assigned from CHAD to the person modeled.

For the nonresidential microenvironments (office, school, store, restaurant, bar, vehicle), PM concentrations are determined using a linear regression equation developed from analysis of concurrent indoor and outdoor PM measurement data available for these microenvironments (Burke et al., 2001):

$$C_{microenvironment} = b^0 + b^1 \times C_{ambient} \quad (4)$$

The outputs from the microenvironmental MENTOR/SHEDS-1A modules link exposure event sequences for each member of the sample population with the corresponding microenvironmental concentration profiles for O_3 and $PM_{2.5}$ during these event sequences.

Step 6: Exposure to O_3 and $PM_{2.5}$ occurs via the inhalation of air containing these constituents; thus, the amount of pollutant delivered to the lung is dependent upon the person's inhalation rate. The process of calculating inhalation rates is as follows:

1. Calculation of ideal body mass (bw), in units of kg, using as inputs the age and gender for each member of the sample population:
$$bw = \exp(Z_{score} \times \log(a) + \log(b)) \quad (5)$$
where Z_{score} is a random factor generated from the standard Normal distribution with mean = 0 and standard deviation = 1, and a, b are age and gender specific regression coefficents available in Burmaster and Crouch (1997).
2. Calculation of a daily basal metabolic rate (DBMR), in units of mJ/day, using the individual's body mass, according to the following age-specific empirical equation available from Schofield (1985)
$$\text{DBMR} = c \times \text{bw} + d + Z_{score} \times e \quad (6)$$
where bw is the body mass, Z_{score} is a random factor generated from the standard Normal distribution with mean = 0 and standard deviation = 1, and c, d, e are age- and gender-specific regression coefficients available from Schofield (1985).
3. Conversion of the DBMR to the basal metabolic rate (BMR) in units of kcal/min:
$$\text{BMR} = \text{DBMR} \times 239(\text{kcal/mJ})/1440(\text{min/day}) \quad (7)$$
4. Calculation of the conversion factor (EETOVO$_2$) between energy expenditure (EE) and oxygen uptake rate (VO$_2$):- for males:
$$\text{EETOVO}_2 = 0.2 + U_{score} \times (0.22 - 0.20) \quad (8)$$
for females:
$$\text{EETOVO}_2 = 0.19 + U_{score} \times (0.21 - 0.19) \quad (9)$$
where U_{score} is a random factor generated from the Uniform distribution (0,1).
5. Calculation of the ventilation rate conversion factor (VQ) between inhalation rate (V_E) and oxygen uptake rate

(VO_2) based on METs (metabolic equivalent of tasks) value, which is a dimensionless number associated with a specific activity. The METs values are provided for each activity event in the USEPA CHAD database (available at the website: www.epa.gov/chadnet1).

$$\text{when METs} \leq 4.5 \quad \text{use} \quad \text{VQ} = 25 + U_{\text{score}} \times 5$$

$$\text{when METs} > 4.5 \quad \text{use} \quad \text{VQ} = 31 + U_{\text{score}} \times 4$$

6. The inhalation rate (V_E) (in units of l/min) is then calculated based on the following equation:

$$V_E = \text{BMR} \times \text{METs} \times \text{EETOVO}_2 \times \text{VQ} \qquad (10)$$

Step 7: A new population-based lung dosimetry model was developed in the present study to calculate the delivered doses for individuals of both genders and of different ages, based on the concepts in HUMTRN (Gallegos and Wenzel, 1984), a subroutine of the BIOTRAN model developed by Los Alamos National Laboratories. The calculated inhalation rate is combined with the associated microenvironmental concentrations to estimate the inhaled dose delivered to the lung for each member of the sample population. In order to estimate the inhaled dose of $PM_{2.5}$, lung deposition of particulate matter is considered for three regions of the lungs: nasal–pharyngeal (NP), tracheobronchial (TB), and pulmonary (P). Three empirical values of deposition fractions for $PM_{2.5}$ are obtained from International Commission on Radiological Protection (ICRP) 1966 database (ICRP, 1994) (and its updated ICRP 1972 database (ICRP, 1996)) to calculate the $PM_{2.5}$ amount deposited in these three regions. The inhaled dose of $PM_{2.5}$ is then the sum of the three deposited $PM_{2.5}$ mass quantities. The inhaled dose of O_3 is simply the total amount of O_3 entering the lung (concentration × inhalation rate), since O_3 is very reactive.

The exposure metric of concern in this study is the 24-h aggregated doses calculated from the sum of event-based doses inhaled by the individual during the exposure event sequence:

$$D_i = \sum_{j=1}^{m} D_{ij} = \sum_{j=1}^{m} (C_{ij} \times T_{ij} \times V_{E_{ij}} \times F_i) \qquad (11)$$

where D_i is the 24-h aggregated dose for individual i (μg), D_{ij} the event-based dose inhaled by individual i during event j, C_{ij} the microenvironmental exposure concentrations (μg/m^3) of $PM_{2.5}$ or O_3 for individual i during event j, T_{ij} the time (h) spent by individual i during activity event j, VE_{ij} the inhalation rate (m^3/h) of individual i during event j, and F_i the deposition fraction of pollutant inhaled for individual i ($F_i = 1$ for O_3). The calculated total 24-h aggregated doses of $PM_{2.5}$ for each individual are also separated into doses due to indoor and outdoor sources, since MENTOR/SHEDS-1A calculates the contributions of outdoor and indoor PM sources to the microenvironmental PM concentration in each activity event using the first and second terms of Equation (3). The calculated total O_3 doses are only due to outdoor sources based on the assumption used for deriving Equation (2). However, since MENTOR/SHEDS-1A keeps track of the time series of microenvironmental exposures, the calculated total O_3 doses are also separated into doses due to time spent indoors and outdoors.

Model Inputs

Inputs to the MENTOR/SHEDS-1A exposure and dose modules include two types of data: microenvironmental data and population/demographic data. Microenvironmental data can be further divided into three categories: outdoor, residential, and nonresidential. Outdoor microenvironmental information was obtained from air quality modeling and the available monitor observations, as previously mentioned. For residential microenvironmental data, input distributions used for the parameters of the indoor residential mass balance equation of $PM_{2.5}$ were obtained from Table 2 of Burke et al. (2001). The same value of air exchange rate (ach) obtained from $PM_{2.5}$ calculation was used to calculate residential O_3 concentrations for each individual also. The input distribution for the residential O_3 decay rates (F_d) was obtained from the study of Weschler et al. (1992). (Alternative distributions for the O_3 decay rates in residences can be found in Lee et al. (1999).) For nonresidential microenvironmental data, input distributions for the parameter used in Equation (3) to calculate $PM_{2.5}$ concentrations were obtained from Table 3 of Burke et al. (2001). For the calculation of O_3 nonresidential concentrations, two distributions were used for the air exchange rate (ach), corresponding to the non-residential (Turk et al., 1989) and vehicle (Hayes, 1991) microenvironments. Two distributions were also used for the O_3 decay rates, corresponding to the non-vehicle (Weschler et al., 1992) and in-vehicle (Hayes, 1989) microenvironments.

Population/demographic data were obtained from available databases such as the US census and human activity surveys (e.g. US Census Housing Survey (US Census Bureau, 2002a) and American Housing Survey (US Census Bureau, 2002b)). Individual diaries of human activity pattern data were obtained from USEPA's CHAD. The CHAD database (McCurdy et al., 2000) has been developed for USEPA's National Exposure Research Laboratory by ManTech Environmental Technologies. It contains over 22,000 person days of activity; all ages and both genders are included, and information regarding every activity undertaken during a day, and lasting for a minute or more, is included in sequential order. Input data on smoking, for determining exposure to environmental tobacco smoke (ETS) in the residence, were obtained from Tables 4–7 of Burke et al. (2001).

Results and discussion

Air Quality Modeling

The study compared CMAQ predictions of ambient pollutant concentrations, including ozone and fine PM, with the corresponding available monitoring data from EPA's AIRS database, and the data collected during the Northeast Oxidant and Particle Study (NE-OPS). Figure 3 shows $PM_{2.5}$ and O_3 time series comparisons of model predictions with NE-OPS measurement data collected by Harvard School of Public Health and Philadelphia Air Management Services, respectively. The maximum, minimum and average of 4 km resolution CMAQ predictions are calculated from nine grid cells in the neighborhood of the monitoring station. The model appears to capture the basic trends during the course of 2 weeks for both $PM_{2.5}$ and O_3. We can see that for O_3 the predictions show excellent agreement with the observation data. $PM_{2.5}$ does not follow the strong diurnal cycles as O_3 does, and we do not see agreements as good as with the O_3 comparisons. Detailed results on the implementation and evaluation of the meteorological and photochemical modeling components of this study are available in Chandrasekar et al. (2001a, 2003a, b, c, d); and Sun et al. (2002, 2003).

A sample "spatial" comparison of CMAQ O_3 results with interpolated fields from observed data, for 1300 hours EDT on 19 July 1999, is shown in Figure 4. Figure 4a shows the CMAQ results for the 4 km resolution grid, at 1300 hours on 19 July 1999. Figure 4b shows the corresponding STRF interpolation results for observed data. Stars in Figure 4b show locations of AIRS monitoring stations from which the data were obtained, and blank areas indicate locations were sufficient data were not available for spatiotemporal estimation. This figure indicates that the spatial distribution patterns of concentrations predicted by CMAQ are consistent with the observed patterns at that particular hour.

Local (Census Tract) and Microenvironmental Concentrations

The STRF and BME methods were used for interpolation of CMAQ predictions and observed ozone concentrations to census tract levels. A total of 482 census tracts in urban Philadelphia were chosen for this case study. Some census tracts, such as those containing airports, railway stations and other industrial areas, were excluded. The excluded census tracts are shown as blank areas in the maps of Figure 5. Interpolations were done at hourly intervals, from 2400 hours EDT on 11 July 1999, to 2300 hours EDT on 24 July 1999. A cross-validation study evaluating the accuracy of the STRF and BME interpolation methods, and comparing the estimates obtained through the two methods, is presented in Georgopoulos et al. (2004a).

The STRF interpolations used 12 nearest neighbors, for both model predictions and observed concentrations. The nearest neighbors were selected through a moving local neighborhood scheme. The data were selected from the estimates/observations 1 h prior to 1 h succeeding the time

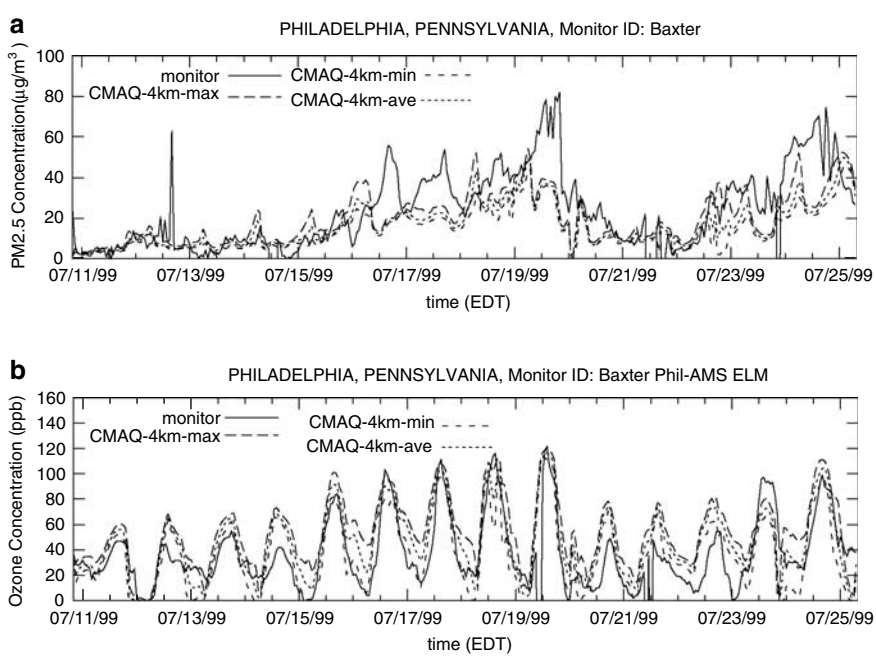

Figure 3. Comparisons of 4 km resolution CMAQ domain (a) PM2.5 and (b) O_3 predictions with observation data for the NE-OPS Baxter site in northern Philadelphia. The monitor data for PM2.5 are taken from the measurements of Harvard SPH during the NE-OPS study and that for O_3 are taken from the measurements of Philadelphia Air Management Services. All concentrations are 1-h averages.

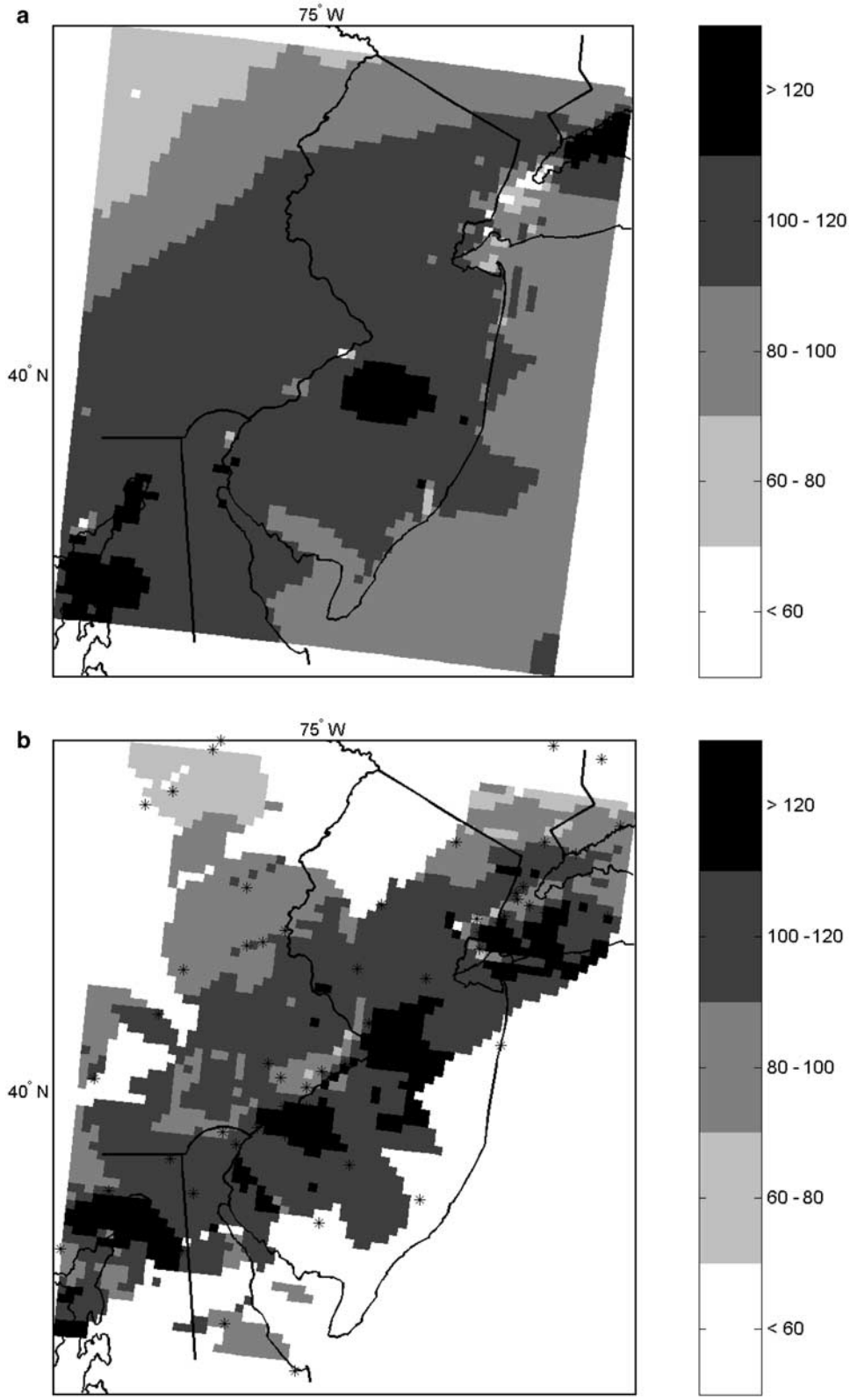

Figure 4. (a) Comparison of CMAQ predicted 1 h averaged O_3 concentrations and (b) interpolated O_3 observed data, at 1300 EDT on 19 July 1999, for the 4-km resolution domain (concentration units are ppb). The CMAQ (Community Multiscale Air Quality) regional photochemical model was used for (a), with emissions processed by SMOKE (Sparse Matrix Operator Kernel Emissions) and meteorological inputs from MM5 (Mesoscale Meterorological Model Version 5). STRF (Spatio-Temporal Random Field) theory algorithms were used for (b).

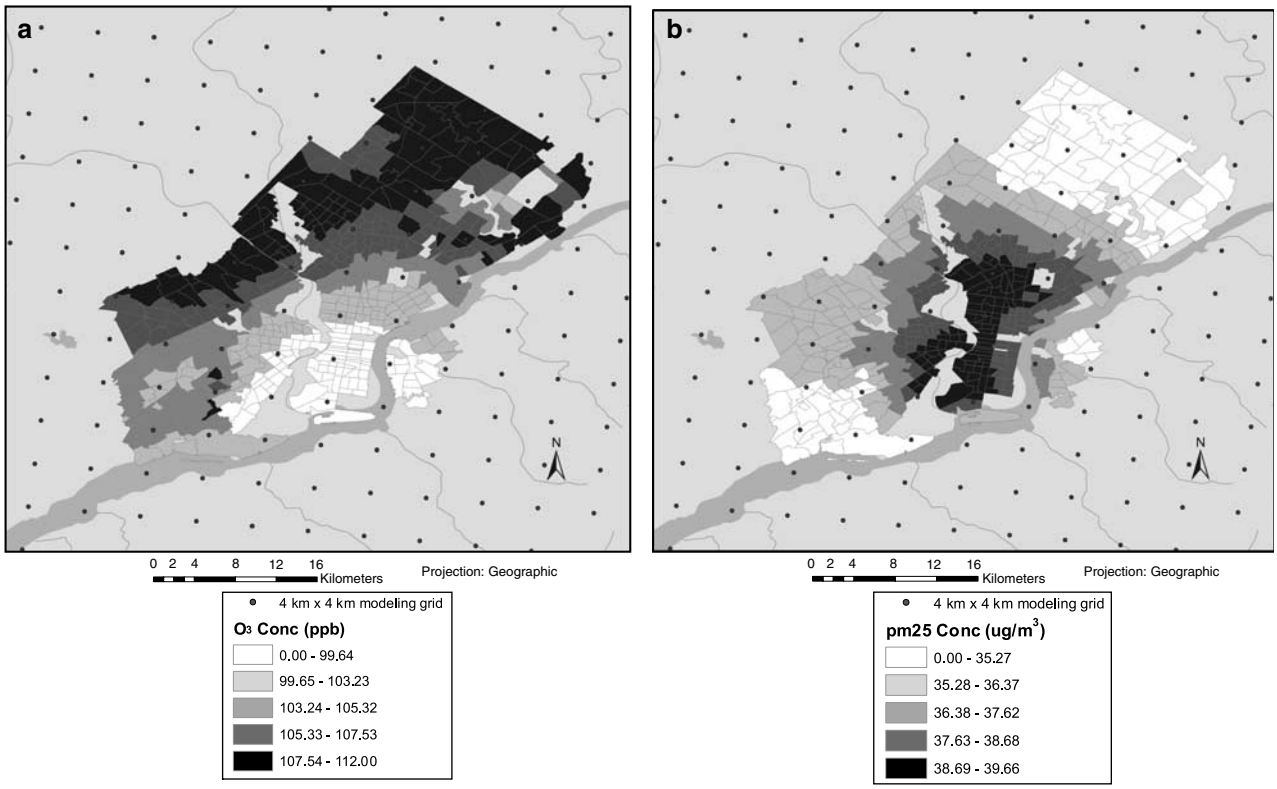

Figure 5. Interpolated outdoor local ambient concentrations for 482 urban Philadelphia census tracts, from CMAQ 1-h averaged predictions at census tract level, using the STRF method, 1300 hours EDT, 19 July 1999. (a) Interpolated O_3 local ambient concentrations, and (b) interpolated $PM_{2.5}$ local ambient concentrations. The grayscale legend scheme represents quantiles of local ambient concentrations. The blank census tracts represent airports and other areas not considered in the study.

instant of estimation. The spatial neighborhood was 5 km for CMAQ predictions, and 30 km for observed data; to include sufficient number of data for the spatiotemporal estimation algorithm. Representative temporal snapshots of interpolated O_3 and $PM_{2.5}$ concentrations, for 1300 hours on 19 July 1999 are presented in Figure 5. While all computations have been performed for the entire 14-day period, representative results are shown for 19 July 1999 (a weekday) for concentrations as well as doses, as this date corresponds to the peak of the episode in the Philadelphia region.

The relationship between outdoor concentrations and calculated microenvironmental concentrations can be seen in the regression calculations presented in Figure 6. For $PM_{2.5}$ the correlation between outdoor concentrations and the 50th percentile of microenvironmental concentrations was stronger than the correlation to the 95th percentiles of microenvironmental concentrations. This indicates that indoor $PM_{2.5}$ sources influence the high end of microenvironmental concentrations. Conversely, for O_3, outdoor concentrations have very strong correlation to both the 50th and 95th percentiles of microenvironmental concentrations, due to lack of indoor sources.

The additional results presented in Georgopoulos et al. (2004a) indicate that the outcomes from the STRF and BME methods are essentially identical, with minor differences at the upper and lower extremes of the concentration range; so, significant inferences cannot be made regarding any differences between the two interpolation methods.

The temporal evolution of the concentrations for the 14 days of interest is shown in Figure 7a and b. The outdoor concentrations over the 482 census tracts were aggregated, and 50th and 95th percentiles at every hour are shown in the time plot. The O_3 outdoor concentrations show marked diurnal fluctuations, which are less evident in the $PM_{2.5}$ concentrations. In both cases, the difference between 50th and 95th percentiles was not very pronounced for the most part. The corresponding total dose percentiles are shown in Figure 7c and d. The 24-h aggregated total dose percentiles for ozone and PM were obtained by lumping together data for all 482 census tracts, for each of the 14 days. It can be seen that the doses do not demonstrate diurnal fluctuations, and the 95th percentiles are significantly higher than the 50th percentiles. This difference is attributed to the influence of activity patterns. However, the daily trend of 95th percentiles of the 24-h aggregated O_3 doses appears to follow the daily trend of outdoor O_3 peak concentrations, which is less evident for the $PM_{2.5}$ case. This is because the total O_3 doses have only contribution from outdoor sources, while the total

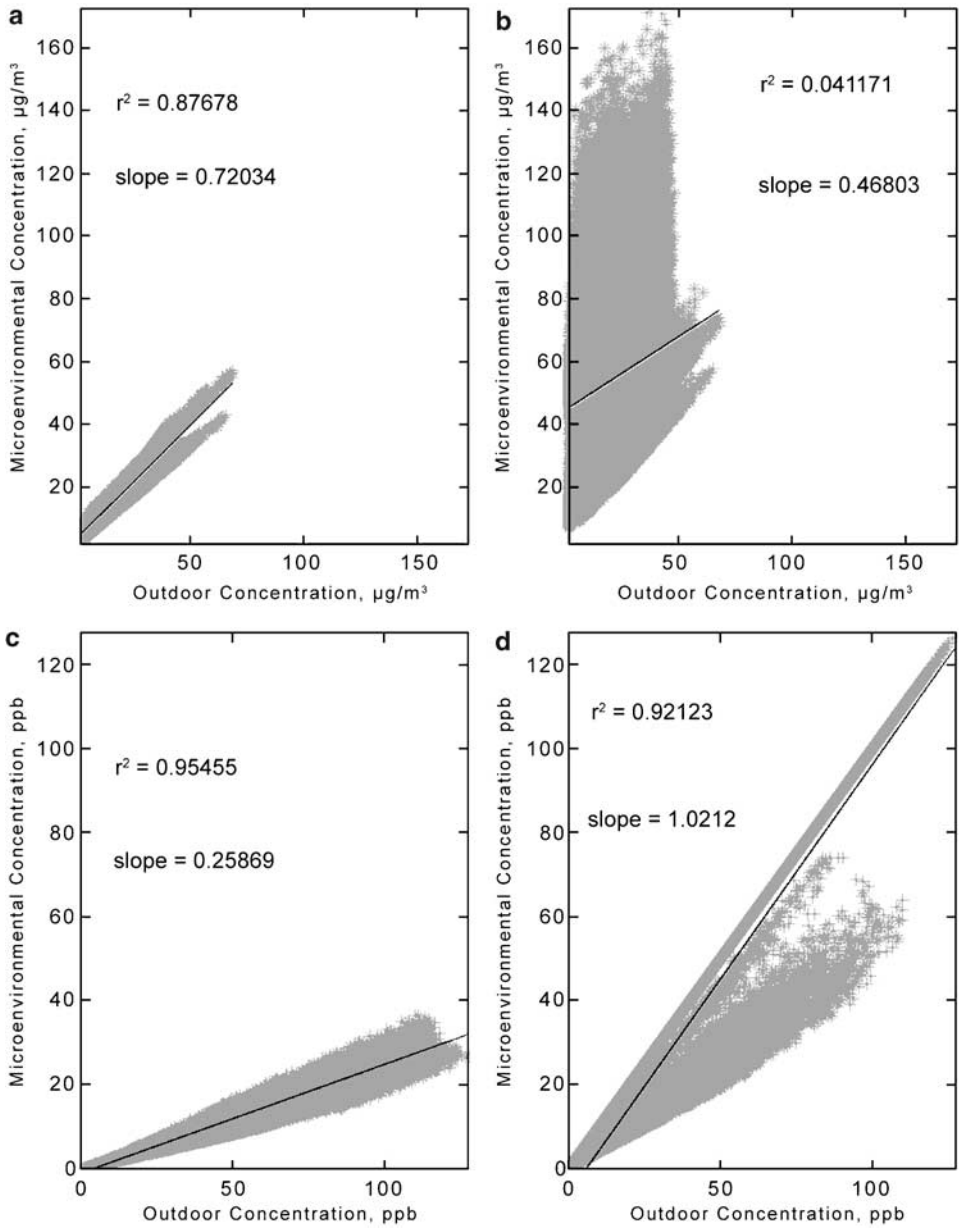

Figure 6. Regression between outdoor and (a) 50th percentile and (b) 95th percentile microenvironmental $PM_{2.5}$ concentrations, 11–24 July 1999. Regression between outdoor and (c) 50th percentile and (d) 95th percentile microenvironmental O_3 concentrations, 11–24 July 1999.

$PM_{2.5}$ doses contain both outdoor and indoor source contributions.

Population Doses

The 24-h aggregated doses for $PM_{2.5}$ and O_3 were estimated using the MENTOR/SHEDS-1A module. For $PM_{2.5}$, estimates were developed for (a) total doses due to all sources; (b) total doses due to indoor sources; and (c) total doses due to outdoor sources. For O_3, estimates were developed for (a) total doses; (b) doses due to time spent outdoors, and (c) doses due to time spent indoors. The difference in classification of doses between $PM_{2.5}$ and O_3 is due to the lack of indoor sources of O_3. MENTOR/SHEDS-1A modules were used to compute population dose estimates.

The relationship between doses and outdoor concentrations was further investigated using multivariate linear regression. The 50th percentiles of the three types of doses for each census tract were regressed to outdoor concentrations at the census tract level. In the case of $PM_{2.5}$, there are significant indoor sources of PM such as cooking, smoking, etc. Hence, while outdoor concentrations were strongly correlated to doses due to outdoor sources, there was no

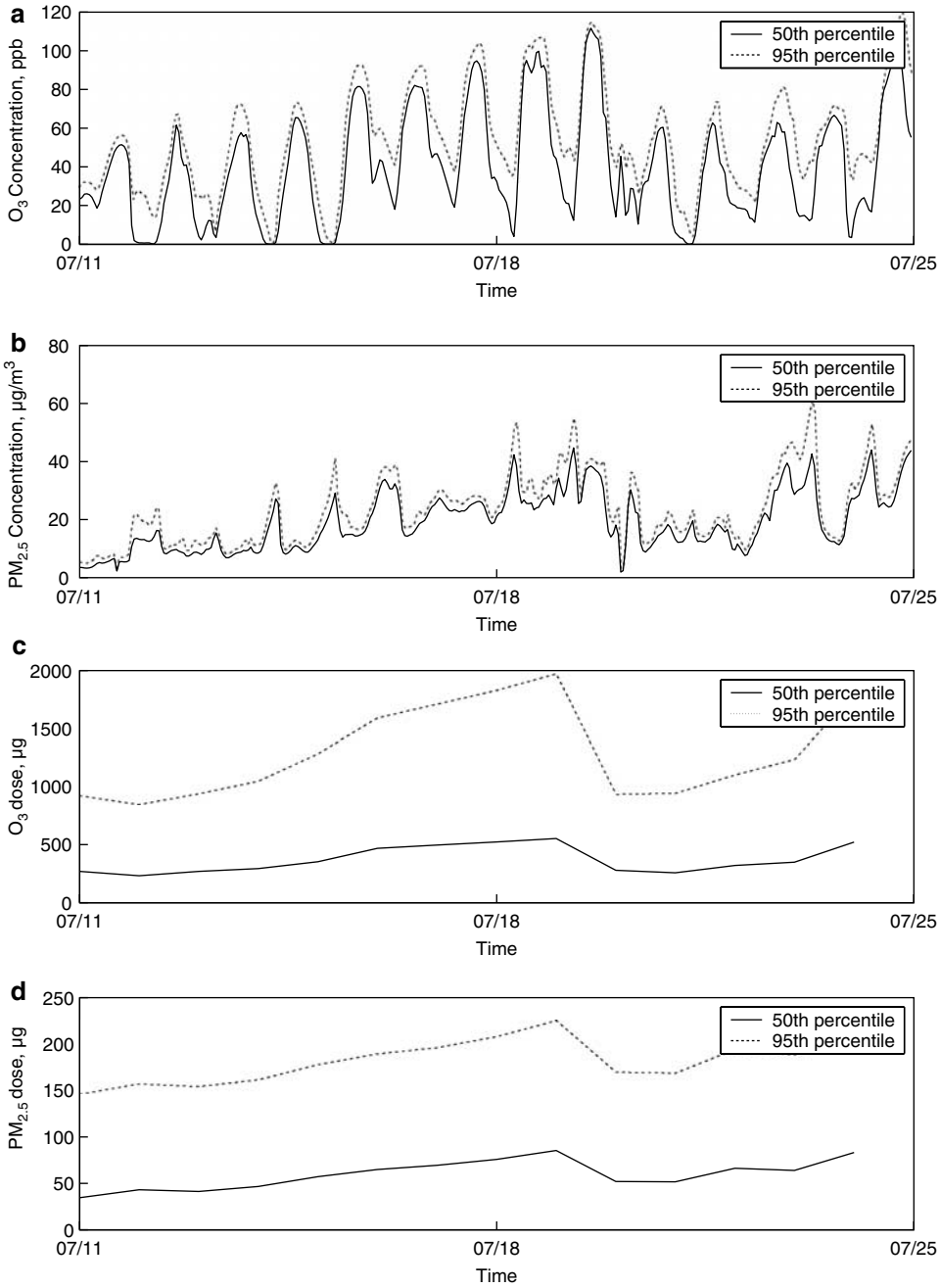

Figure 7. Time plots of outdoor O_3 and $PM_{2.5}$ local ambient concentrations and corresponding total doses, for the selected 482 census tracts in urban Philadelphia; 11–24 July 1999: (a) O_3 outdoor local ambient concentrations, (b) $PM_{2.5}$ outdoor local ambient concentrations, (c) O_3 total dose from time spent indoors as well as outdoors, and (d) $PM_{2.5}$ total dose from indoor and outdoor sources.

correlation between outdoor concentrations and doses due to indoor sources (Figure 8). The relationship between doses and tobacco smoke was also investigated. Figure 8d shows that the regression between number of smokers in a census tract and total $PM_{2.5}$ dose explained about 15% of the variation in the doses due to indoor sources where number of smokers was used as an input variable. Figure 9 shows the corresponding relationship between outdoor O_3 concentrations and 95th percentiles of doses. Since no indoor sources of O_3 were considered, the correlation is strong between outdoor O_3 concentrations and doses due to time spent indoors as well as doses due to time spent outdoors.

The cumulative distributions of the 24-h aggregated O_3 and $PM_{2.5}$ doses for 19 July 1999 are shown in Figure 10. The total $PM_{2.5}$ dose distribution has a median of 85.3 μg,

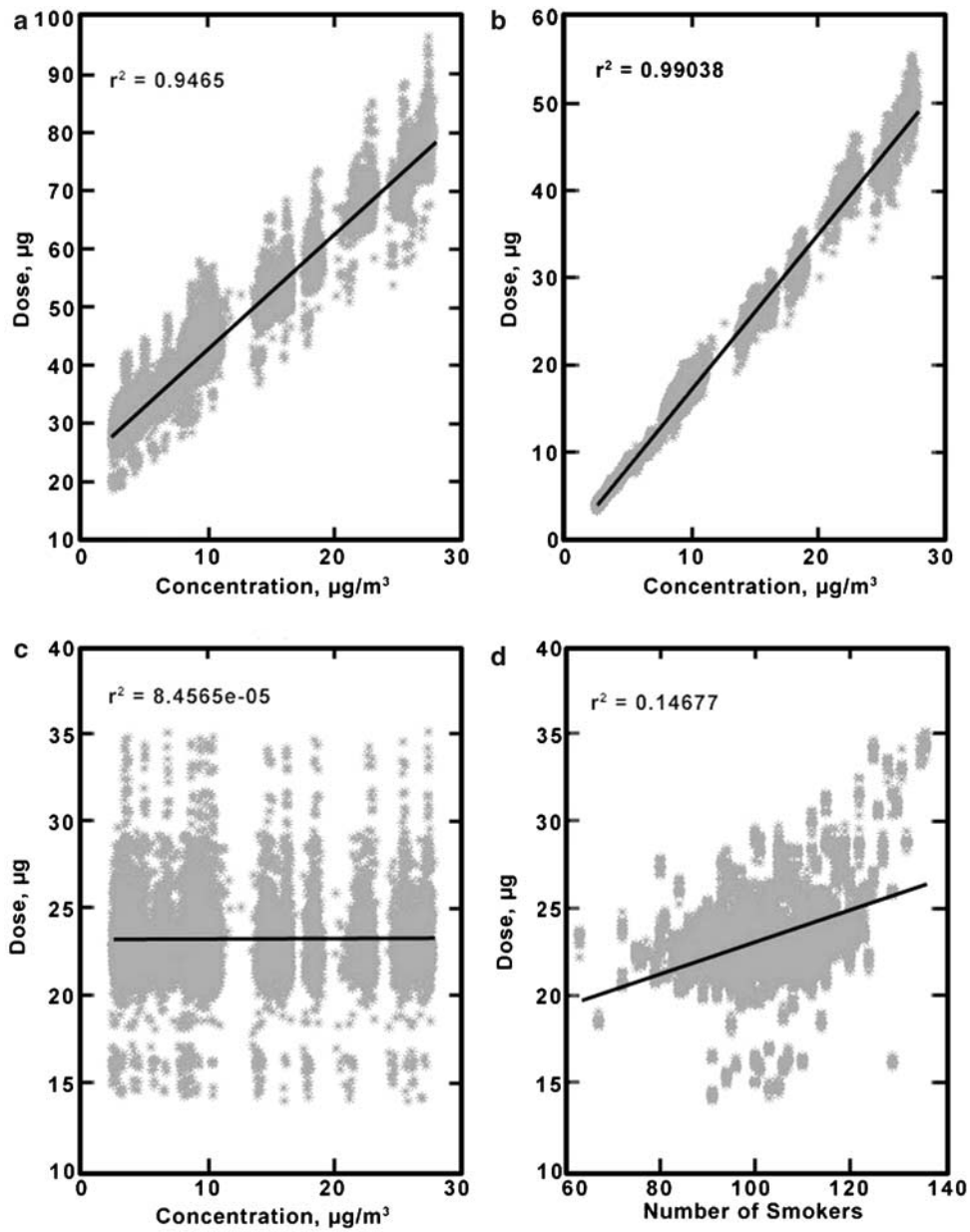

Figure 8. Regression between 50th percentiles of $PM_{2.5}$ doses and outdoor local ambient concentrations in each census tract, computed using STRF interpolation of CMAQ predictions: (a) outdoor local ambient concentrations to total doses; (b) outdoor local ambient concentrations to doses due to outdoor sources; (c) outdoor local ambient concentrations to doses due to indoor sources; and (d) number of smokers per census tract to doses due to indoor sources. All results are for the 482 census tracts in urban Philadelphia.

with the upper 5% of the simulation population having dose levels greater than 224.7 μg. Comparing with the distribution statistics of $PM_{2.5}$ doses due to indoor or outdoor sources separately, it is found that indoor sources dominate the contributions to the total doses for the upper five percentiles. Specifically, the maximum value of total $PM_{2.5}$ doses has 98.6% contribution from indoor sources, which are mostly from ETS. To further investigate the ETS contribution to the $PM_{2.5}$ doses due to indoor sources for the upper 5% of the simulation population, the ratio of $PM_{2.5}$ doses due to ETS divided by the doses due to all indoor sources was calculated for each individual of the upper 5% of the population. It was found that 65% of these high-end exposed population have over 72% contribution from ETS among all indoor sources.

The total O_3 dose distribution has a median of 552 μg, with the upper 5% of the simulation population having dose levels greater than 1,968 μg. Comparing with the distribution

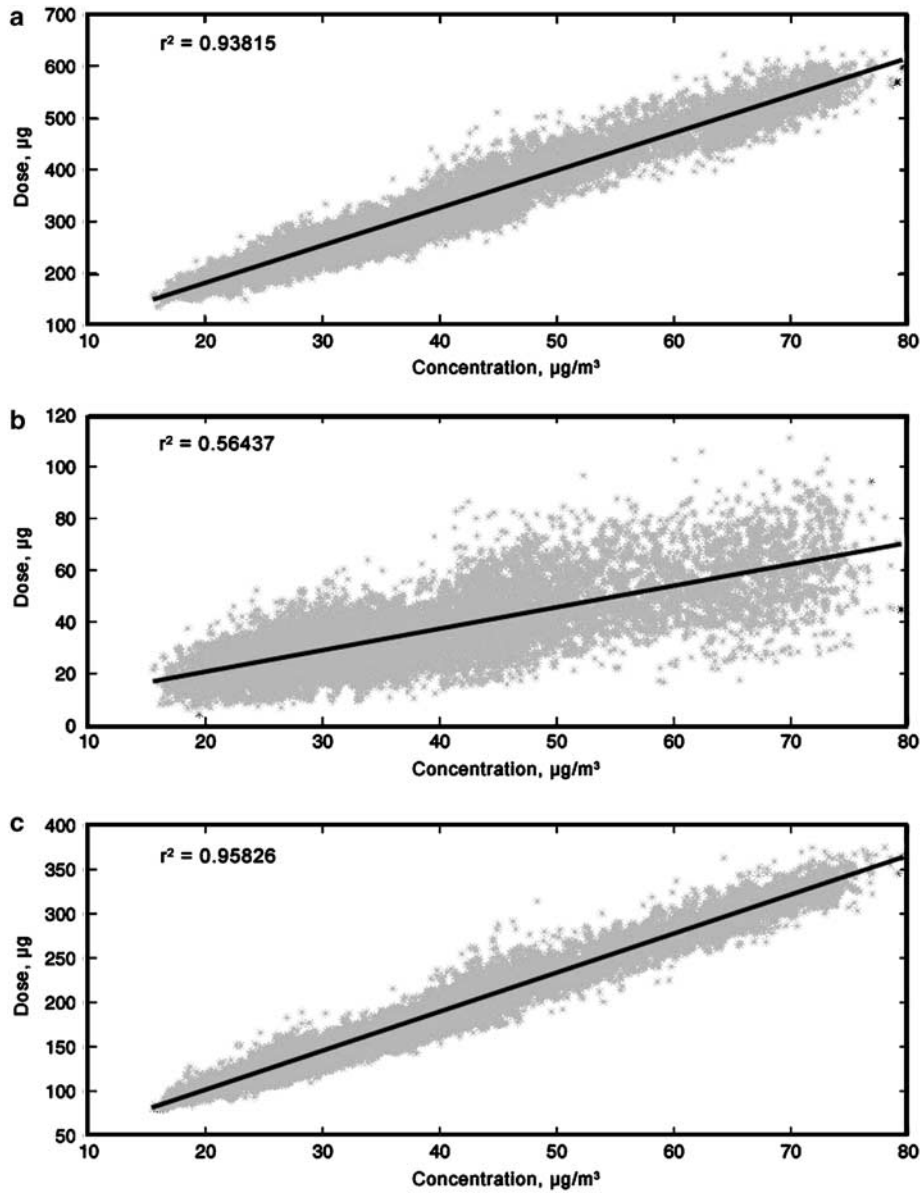

Figure 9. Regression between 50th percentiles of O_3 doses and outdoor concentrations in each census tract, computed using STRF interpolation of CMAQ predictions: (a) outdoor concentrations to total doses; (b) outdoor concentrations to doses due to time spent outdoors; and (c) outdoor concentrations to doses due to time spent indoors. All results are for 482 census tracts in urban Philadelphia, for 11–24 July 1999.

statistics of O_3 doses due to time spent indoors or outdoors separately, it is found that dose due to time spent outdoors dominates the contributions to the total doses for the upper five percentiles. Specifically, the maximum value of total O_3 doses has 97% contribution from doses due to time spent outdoors.

Co-occurrence Analysis

To facilitate comparison of patterns in $PM_{2.5}$ and O_3 occurrences, the modeled concentrations were normalized by the daily standards for both parameters — the 80 ppb level for the maximum daily 8-h running average for O_3, and the 65 $\mu g/m^3$ daily average for $PM_{2.5}$. The O_3 concentrations are normalized as

$$n_{O_3} = \frac{(a8_{O_3} - 80)}{80}$$

where $a8_{O_3}$ is the daily maximum value of 8-h running averages of observed O_3 concentrations in units of ppb. The $PM_{2.5}$ concentrations are normalized as

$$n_{PM_{2.5}} = \frac{(a_{PM_{2.5}} - 65)}{65}$$

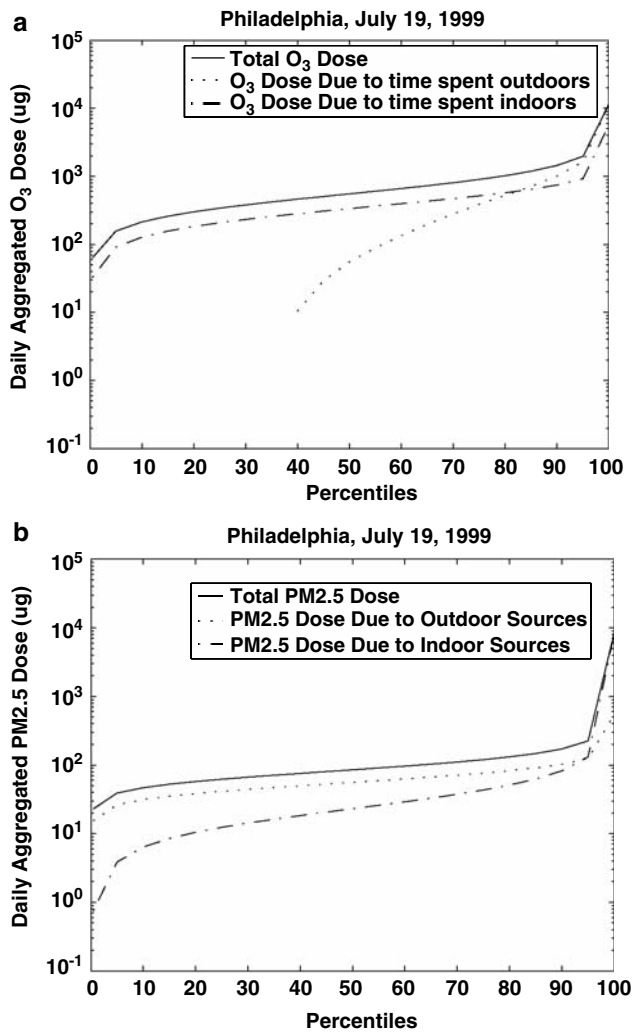

Figure 10. The cumulative distributions of the 24-h aggregated (a) O_3 and (b) $PM_{2.5}$ doses for 19 July 1999.

where $a_{PM_{2.5}}$ is the daily (24-h) averaged $PM_{2.5}$ concentration in units of $\mu g/m^3$. The ratios of normalized values were then computed for each census tract, for each day, as

$$r_n = \frac{n_{O_3}}{n_{PM_{2.5}}}$$

The maps of ratios provide a means of examining the spatial distribution of $PM_{2.5}$ and O_3 co-occurrence patterns; this is straightforward when $PM_{2.5}$ values do not exceed the $65\,\mu g/m^3$ daily average standard as is the case in the present study. If this assumption is satisfied, then $n_{PM_{2.5}}$ will always be negative. Positive values of n_{O_3} indicate locations and magnitude of O_3 exceedances. The ratio r_n will be positive when both parameters do not exceed their respective standards, and it will be negative when O_3 exceeds its standard but $PM_{2.5}$ does not. Large negative values indicate instances where O_3 exceedances are the dominant phenomenon (i.e. O_3 pollution is the predominant problem). Likewise, large positive values should indicate locations and instances where O_3 pollution is not a problem. The ratios between normalized O_3 and $PM_{2.5}$ concentrations, for 16 and 19 July are presented in Figure 11a and b, for STRF interpolations. The maps show identical patterns for 16 and 19 July. The ratios indicate that the pollution episode was dominated by high O_3 values throughout the Philadelphia area on 19 July, but less so on 16 July.

Conclusions

This study demonstrated the feasibility of developing population exposure assessments, for O_3 and PM, using an integrated and mechanistically consistent source-to-dose framework, provided by the combined MENTOR/SHEDS-1A system. The case study presented here utilized USEPA's multiscale photochemical modeling framework (Models-3/CMAQ) with MM5 for meteorological modeling, SMOKE for emissions modeling, and CHAD for characterizing time/activity patterns, in combination with the MENTOR/SHEDS-1A system.

It was found that 50th percentiles of microenvironmental concentrations of $PM_{2.5}$ and O_3 were significantly correlated with census-tract level outdoor concentrations, respectively. Furthermore, 95th percentiles of O_3 microenvironmental concentrations were also strongly correlated with outdoor concentrations, which was not the case for $PM_{2.5}$. This indicates that non-ambient $PM_{2.5}$, but rather indoor sources influence the high end of $PM_{2.5}$ microenvironmental concentrations. By further examining the distribution statistics of the 24-h aggregated $PM_{2.5}$ and O_3 doses, it was found that indoor $PM_{2.5}$ sources dominate the contributions to the total $PM_{2.5}$ doses for the upper five percentiles, while the total O_3 doses have only the contribution from outdoor sources due to the lack of indoor sources of O_3 production. Among various $PM_{2.5}$ indoor sources, ETS contributed the most to the high-end of 24-h aggregated $PM_{2.5}$ doses. The O_3 doses due to time spent outdoors dominate the contributions to the total O_3 doses for the upper five percentiles. The spatial distributions of $PM_{2.5}$ and O_3 co-occurrence patterns were also examined in this study. It was shown that O_3 exceedances were the dominant phenomenon for some days of the 2-week pollution episode, while $PM_{2.5}$ values did not exceed the $65\,\mu g/m^3$daily average standard during the whole period.

It is important to note that the MENTOR/SHEDS-1A system developed in this study provides a consistent framework for characterizing pollutant sources, exposure factors, and human activity patterns, in conducting population exposure assessments simultaneously for multiple pollutants. In this study, the same physical and chemical assumptions

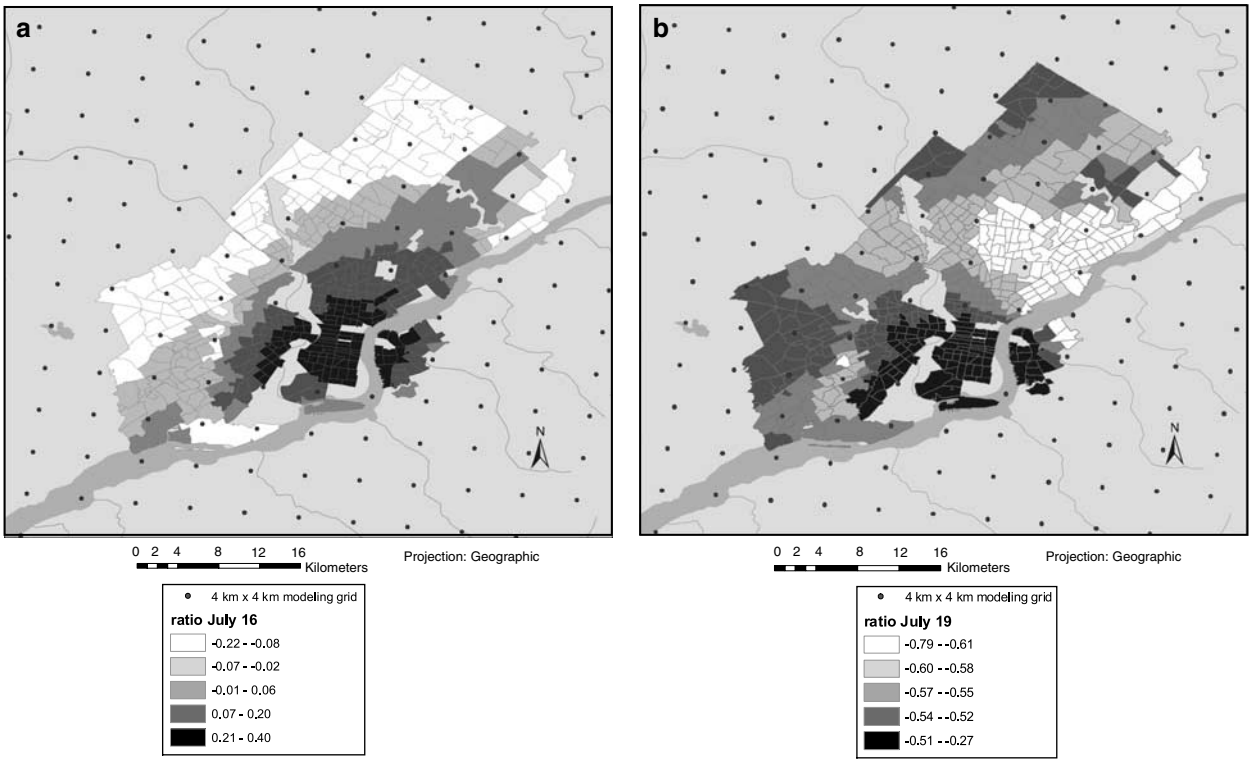

Figure 11. Ratio of normalized O_3 local ambient concentrations to normalized $PM_{2.5}$ local ambient concentrations, for (a) 16 July 1999 and (b) 19 July 1999, over Urban Philadelphia. Census tract level interpolations were done using the BME method. All results are for 482 census tracts in urban Philadelphia, and the grayscale legend scheme represents quantiles of the normalized local ambient concentration distribution. Blank areas are census tracts that were not included in the study.

were used in air quality modeling for obtaining both $PM_{2.5}$ and O_3 outdoor concentrations; the same residential indoor/outdoor air exchange rate was used to calculate both $PM_{2.5}$ and O_3 microenvironmental concentrations for each individual of the simulated population; the same activity diary and related inhalation rates were used to calculate the inhaled doses of both $PM_{2.5}$ and O_3 for each individual of the simulated population. These modeling features of MENTOR/SHEDS-1A system are the primary distinctions from previous studies of population exposure assessment, where different exposure factors and activity patterns may be used for different pollutants.

The MENTOR/SHEDS-1A system inherits the limitations and data needs of the previous SHEDS study (Burke et al., 2001) regarding the calculations of residential and non-residential $PM_{2.5}$ microenvironmental concentrations. Due to insufficient information, the calculations of residential and non-residential O_3 microenvironmental concentrations also have similar limitations and data needs as in the $PM_{2.5}$ case. The MENTOR/SHEDS-1A system improves the time resolution of characterizing outdoor pollutant concentrations from 12 h of the previous SHEDS study to 1 h. State-of-the-art interpolation methods were used to provide the link between photochemical model predictions (or monitored air quality concentrations) and local (census tract level) outdoor concentrations to be used as input for population exposure models. The use of alternative methods for developing outdoor concentrations at the local level, and the employment of two different geostatistical methods, also served to evaluate the photochemical model predictions, interpolation method estimates, and population exposure model results for doses. The MENTOR/SHEDS-1A system is also equipped with the capability of estimating intake dose based on activity levels and inhalation/exhalation rates, which completes the source-to-dose modeling sequence. Future work will focus on characterization of commuting patterns for use in population exposure/dose assessment as well as on extending the MENTOR/SHEDS-1A applications to other areas in the US.

Acknowledgements

The support for this work has been provided primarily by the USEPA funded Center for Exposure and Risk Modeling (CERM) at EOHSI (EPAR-827033); and the State of New Jersey Department of Environmental Protection (NJDEP) funded Ozone Research Center (ORC) at EOHSI. Additional funding has been provided by the USEPA funded NorthEast Oxidant and Particle Study (NE-OPS) University

Consortium (EPA-TPSU-UMDNJ-826373-14), by EPA STAR Grant No. R826768-01, by the NIEHS Center for Environmental Health Sciences at EOHSI (Grant No. P01 ES11256-01), and by the American Chemistry Council (Grant No. 2488). We thank Professors George Christakos and Mark Serre from the University of North Carolina at Chapel Hill for providing access to the BMELIB software library and for their helpful suggestions in implementing BMELIB programs; and Anatharaman Chandrasekar, Pamela Shade, Srinivas Bandi, Hao-Chen Tan, Yu-Ching Yang and Linda Everett of the Computational Chemodynamics Laboratory for their helpful contributions to this study. We are solely responsible for errors, if any, in the BMELIB results and the accompanying analysis presented in this report.

References

Burke J.M., Zufall M.J., and Ozkaynak H. A population exposure model for particulate matter: case study results for $PM_{2.5}$ in Philadelphia, PA. *J Expos Anal Environ Epidemiol* 2001: 11: 470–489.

Burmaster D.E., and Crouch E.A.C. Lognormal distributions for body weight as a function of age for males and females in the United States, 1976–1980. *Risk Anal* 1997: 17: 499–505.

Chandrasekar A., Philbrick C.R., Clark R., Doddridge B., and Georgopoulos P.G. Evaluating the performance of a computationally efficient MM5/CALMET system for developing meteorological inputs to air quality models. *Atmos Environ* 2003a: 37: 3267–3276.

Chandrasekar A., Philbrick C.R., Clark R., Doddridge B., and Georgopoulos P.G. A large-eddy simulation study of the convective boundary layer over Philadelphia during the 1999 summer NE-OPS campaign3. *Environ Fluid Mech* 2003b: 3: 305–329.

Chandrasekar A., Philbrick C.R., Doddridge B., Clark R., and Georgopoulos P.G. Evaluation of mesoscale model fifth generation (MM5) applications with NE-OPS 1999 upper air data: a comparative study of MM5 simulations with aircraft, wind profiler, lidar, tethersonde and RASS NE-OPS data over Philadelphia, PA During a 1999 summer episode (final draft). Interim Technical Report CCL-ITR-2001:ORC.NEOPS-01, 2001a.

Chandrasekar A., Philbrick C.R., Doddridge B., Clark R., and Georgopoulos P.G. Evaluation of regional atmospheric modeling system (RAMS) applications with NE-OPS 1999 upper air data: a comparison study of RAMS simulations with aircraft, wind profiler, lidar, tethered balloon and RASS NE-OPS data over Philadelphia, PA during a 1999 summer episode (final draft). Interim Technical Report CCL-ITR-2001:ORC.NEOPS-02, 2001b.

Chandrasekar A., Philbrick C.R., Doddridge B., Clark R., and Georgopoulos P.G. A comparative study of prognostic MM5 meteorological model with aircraft, wind profiler, lidar, tethered balloon and RASS data over Philadelphia during a 1999 summer episode. *Environ Fluid Mech* 2003c; in press.

Chandrasekar A., Philbrick C.R., Doddridge B., Clark R., and Georgopoulos P.G. An evaluation study of RAMS simulations with aircraft, wind profiler, lidar, tethered balloon and RASS data over Philadelphia during a 1999 summer episode. *Atmos Environ* 2003d: 37: 4973–4984.

Christakos G. On certain classes of spatiotemporal random fields with applications to space–time data processing. *IEEE Trans Systems Man Cybernet* 1991: 21: 861–875.

Clench-Aas J., Bartonova A., and Bohler T. Air pollution exposure monitoring and estimating Part I. Integrated air quality monitoring system. *J Environ Monit* 1999: 1: 313–319.

Douglas S.G., Hudischewskyj A.B., and Gorsevski V. Use of the UAM-V modeling system as an air quality planning tool and for examining heat island reduction strategies. Proceedings of the ACEEE 2000 Summer Study on Energy Efficiency in Buildings. American Council for an Energy Efficient Economy, 2000, pp. 9.83–9.92.

ENVIRON. *Users Guide — Comprehensive Air Quality Model With Extensions (CAMx) Version 3.10*. ENVIRON International Corp., Novato, CA, 2002.

Freijer J.I., Bloemen H.J.T., de Loos S., Marra M., Rombout P.J.A., Steentjes G.M., and van Veen M.P. Modelling exposure of the Dutch population to air pollution. *J Hazard Mater* 1998: 61: 107–114.

Gallegos A.F., and Wenzel W.J., HUMTRN: Documentation and Verification for an ICRP-Based Age- and Sex-Specific Human Simulation Model for Radionuclide Dose Assessment, Los Alamos National Laboratory, Los Alamos, NM, 1984.

Georgopoulos P.G., and Lioy P.J. Conceptual and theoretical aspects of human exposure and dose assessment. *J Expos Anal Environ Epidemiol* 1994: 4: 253–285.

Georgopoulos P.G., and Lioy P.J. MENTOR-based OPERAS (particle exposure and risk analysis systems): an overview. Annual Conference of the International Society of Exposure Analysis, Monterey, CA, 2000.

Georgopoulos P.G., Wang S.W., Vyas V.M., Sun Q., Chandrasekar A., Shade P., Vedantham R., Burke J., McCurdy T., and Özkaynak H. A source-to-dose assessment of population exposures to fine PM, ozone and air toxics in Philadelphia, PA, during a 1999 summer episode. A case study employing MENTOR/SHEDS-1A. Technical Report CERM:2004-01, Prepared for USEPA (Revised Draft submitted for agency review), 2004a.

Georgopoulos P.G., Wang S.W., Yang Y.C., Xue J., Zartarian V., McCurdy T., and Özkaynak H. Assessing multimedia/multipathway exposures to arsenic using a mechanistic source-to-dose modeling framework. Case studies employing MENTOR/SHEDS-4M. Technical Report CERM: 2004-02, Prepared for USEPA (Revised Draft submitted for agency review), 2004b.

Glen G. *User's Guide for the APEX3 Model*. ManTech Environmental Technology Inc. (prepared for U.S. EPA), Research Triangle Park, NC, 2002.

Grell G.A., Dudhia J., and Stauffer D.R. A description of the fifth-generation Penn State/NCAR mesoscale model (MM5). NCAR Technical Note TN-398 + STR, National Center for Atmospheric Research, Boulder, CO, 1994.

Guinnup D., and Collom B., Telling the OTAG Ozone Story with Data — Final Report, Vol. I, Ozone Transport Assessment Group (OTAG) Air Quality Analysis Workgroup, 1997.

Hayes S.R. Estimating the effect of being indoors on total personal exposure to outdoor air pollution. *J Air Pollut Control Assoc* 1989: 39: 1453–1461.

Hayes S.R. Use of an indoor air quality model (IAQM) to estimate indoor ozone levels. *J Air Waste Manage Assoc* 1991: 41: 161–170.

Houyoux M.R., and Vukovich J.M. *Updates to the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System and Integration with Models-3, The Emission Inventory: Regional Strategies for the Future*. Air & Waste Management Association, Raleigh, NC, 1999.

ICRP. *Publication 66: Human Respiratory Tract Model for Radiological Protection*. Annals of the ICRP Vol. 24/1-3, Oxford, UK, 1994.

ICRP. *Publication 72: Age-dependent Doses to the Members of the Public from Intake of Radionuclides: Part 5, Compilation of Ingestion and Inhalation Coefficients*. Annals of the ICRP Vol. 26/1, Oxford, UK, 1996.

Johnson T.R., Capel J.E., McCoy M.D., and McCurdy T. Estimation of ozone exposures experienced by urban residents using an enhanced version of PNEM. In: J.J. Vostal (Ed.), *Tropospheric Ozone: Critical Issues in the Regulatory Process*. Air and Waste Management Association, Pittsburgh PA, 1996, pp. 209–220.

Lee K., Vallarino J., Dumyahn T., Ozkaynak H., and Spengler J.D. Ozone decay rates in residences. *J Air Waste Manage Assoc* 1999: 49: 1238–1244.

Lioy P.J. The analysis of total human exposure for exposure assessment: a multidiscipline science for examining human contact with contaminants. *Environ Sci Technol* 1990: 24: 938–945.

Lurmann F., Colome S.D., and Hogo H. Modeling current and future human exposure to ozone in Southern California. In: R.L. Berglund (Ed.), *Tropospheric Ozone and the Environment II*. Air and Waste Management Association, Pittsburgh, PA, 1992, pp. 725–745.

Lurmann F.W., and Korc M.E. Characterization of human exposure to ozone and PM10 in the San Francisco Bay area. Final Report. STI 93150 1416 FR, Bay Area Air Quality Management District, San Francisco, CA, 1994.

McCurdy T., Geln G., Smith L., and Lakkadi Y. The National Exposure Research Laboratory's consolidated human activity database. *J Expos Anal Environ Epidemiol* 2000: 10: 566–578.

McCurdy T.R. Human exposure to ambient ozone. In: D.J. McKee (Ed.), *Tropospheric Ozone: Human Health and Agricultural Impacts*. Lewis Publishers, Boca Raton, FL, 1994, pp. 85–127.

Morris R., Yarwood G., Emery C., and Wilson G. Modeling PM chemistry and PMCAMx update. Presented at Midwest RPO Modeling Meeting. ENVIRON International Corporation, Des Plaines, IL, 2002.

NCAR. *PSU/NCAR Mesoscale Modeling System Tutorial Class Notes and User's Guide*. National Center for Atmospheric Research, Mesoscale and Microscale Meteorology Division, CO, 1999.

NOAA. National Climatic Data Center Web Site (http://www.ncdc.noaa.gov/oa/ncdc.html). National Oceanic and Atmospheric Administration, 2003.

Odman M.T., and Ingram C.L. Multiscale Air Quality SImulation Platform (MAQSIP): source code documentation and validation. ENV-96TR002-v1.0, MCNC, Research Triangle Park, NC, 1996.

Ott W.R. Concepts of human exposure to air pollution. *Environ Int* 1982: 7: 179–196.

Ott W.R. Total human exposure. *Environ Sci Technol* 1985: 19: 880–885.

Özkaynak H., Xue J., Spengler J., Wallace L., Pellizzari E., and Jenkins P. Personal exposure to airborne particles and metals: results from the Particle TEAM study in Riverside, California. *J Expos Anal Environ Epidemiol* 1996: 6: 57–78.

Philbrick C.R. Investigations of factors determining the occurrence of ozone and fine particles in Northeastern USA. Proceedings of Symposium on Measurement of Toxic and Related Air Pollutants. Air and Waste Management Association, 1999, pp. 248–260.

Philbrick C.R., Ryan W.F., Clark R.D., Doddridge B.G., Dickerson R.R., Koutrakis P., Allen G., McDow S.R., Rao S.T., Hopke P.K., Eatough D.J., Dasgupta P.K., Tollerud D.J., Georgopoulos P., Kleinman L.I., Daum P., Nunnermacker L., Dennis R., Schere K., McClenny W., Gaffney J., Marley N., Coulter R., Fast J., Doren C., and Mueller P.K. Overview of the NARSTO-NE-OPS program. Proceedings of the Fourth Conference on Atmospheric Chemistry: Urban, Regional, and Global-Scale Impacts of Air Pollutants. American Meteorological Society, Orlando, FL, 2002, pp. 107–114.

Rifai H.S., Hopkins L., and Mehdizadeh F. Developing human exposure estimates for indoor/outdoor air pollution in Houston, *Environmental Institute of Houston — 1999 Annual Report*. Environmental Institute of Houston, Houston, TX, 2000.

Rosenbaum A. *The HAPEM4 User's Guide Hazardous Air Pollutant Exposure Model, Version 4*. USEPA Office of Air Quality Planning and Standards, Research Triangle Park, NC, 2002.

Schofield W.N. Predicting basal metabolic rate, new standards, and review of previous work. *Hum Nutr Clin Nutr* 1985: 39C: 5–41.

Scire J.S., Robe F.R., Fernau M.E., and Yamartino R.J. *A User's Guide for the CALMET Meteorological Model (Version 5)*. Earth Tech, Inc., Concord, MA, 2000.

Serre M., and Christakos G. Modern geostatistics: computational BME in the light of uncertain physical knowledge — the Equus Beds study. *Stochast Environ Res Risk Assess* 1999: 13: 1–26.

Stockwell W.R., Kirchner F., Kuhn M., and Seefeld S. A new mechanism for regional atmospheric chemistry modeling. *J Geophys Res* 1997: 102: 25847–25879.

Stockwell W.R., Middleton P., Chang J.S., and Tang X. The second generation regional acid deposition model chenical mechanism for regional air quality modeling. *J Geophys Res* 1990: 95, 16,343–16,368.

Strum M., Mason R., and Linderman D. *User's Guide for the Emissions Modeling System for Hazardous Air Pollutants (EMS-HAP) Version 2.0*. EPA 454/B-02-001, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Emissions, Monitoring and Analysis Division, Research Triangle Park, NC, 2002.

Sun Q., Chandrasekar A., and Georgopoulos P.G. Evaluation of models-3/CMAQ with AIRS and NE-OPS data for a 1999 summer episode with focus on New Jersey and the Philadelphia Area (Draft). Technical Report ORC:2002-01, Prepared for the NJ Department of Environmental Protection, 2002.

Sun Q., and Georgopoulos P.G. Comparative evaluation of the performance of the CB4 and RADM2 chemical mechanisms in models-3/CMAQ with focus on the prediction of ozone and $PM_{2.5}$ levels in New Jersey and the Philadelphia Area. Technical Report ORC:2002-02, Prepared for the NJ Department of Environmental Protection, 2002.

Sun Q., Chandrasekar A., and Georgopoulos P.G. Comparative evaluation of photochemical model predictions using the NE-OPS 1999 field data. *Technical Report ORC:2003-01*. Ozone Research Center, Piscataway, NJ, 2003.

Turk B.H., Grimsrud D.T., Brown J.T., Geisling-Sobotka K.L., Harrison J., and Prill R.J. Commercial building ventilation rates and particle concentrations. *ASHRAE Trans* 1989: 95: 422–433.

US Census Bureau. 1990 Census of Population, General Population Characteristics, United States. 1990 CP-1-1, Washingston, DC, 1992.

US Census Bureau. 2000 Census of Population and Housing, Summary Population and Housing Characteristics, United States. PHC-1-1, Washington DC, 2002a.

US Census Bureau. Current Housing Reports, Series H150/01, American Housing Survey for the United States: 2001, US Government Printing Office, Washington DC, 2002b.

USEPA. Final Guidelines for Exposure Assessment. EPA/600Z-92/001, US EPA, National Center for Environmental Assessment, 1992.

USEPA. Exposure Factors Handbook, US EPA, National Center for Environmental Assessment, Cincinnati, OH, 1997.

USEPA. Total Risk Integrated Methodology, TRIM.Expo.: Technical Support Document. EPA-453/D-99-001, USEPA Office of Air Quality Planning and Standards, Office of Air and Radiation, Research Triangle Park, NC, 1999a.

USEPA. *User Manual for the EPA Third-Generation Air Quality Modeling System (Models-3 Version 3.0)*. EPA-600/R-99/055, USEPA, Office of Research and Development, Washington, DC, 1999b.

USEPA. National Air Quality and Emissions Trends Report, 1998. EPA 454/R-00-003, US EPA Office of Air Quality Planning and Standards, Research Triangle Park, NC, 2000.

USEPA. AIRS User's Guide. USEPA Office of Air Quality Planning and Standards, 2001a.

USEPA. Procedures Document for National Emission Inventory, Criteria Air Pollutants 1985–1999. EPA-454/R-01-008, US EPA, Office of Air Quality Planning and Standards, Research Triangle Park, NC, 2001b.

USEPA. *User Guide: Air Quality System, USEPA*. Office of Air Quality Planning and Standards, Research Triangle Park, NC, 2002.

USEPA. Report on the Model SHEDS-MENTOR-1A (http://cfpub.epa.gov/crem/crem_report.cfm?deid = 75917), USEPA Council for Regulatory Environmental Modeling (CREM), Washington DC, 2004a.

USEPA. Report on the Model SHEDS-MENTOR-4M (http://cfpub.epa.gov/crem/crem_report.cfm?deid = 75918), USEPA Council for Regulatory Environmental Modeling (CREM), Washington DC, 2004b.

Vyas V.M., and Christakos G. Spatiotemporal analysis and mapping of sulfate deposition data over eastern USA. *Atmos Environ* 1997: 31: 3623–3633.

Walko R.L., and Tremback C.J. *RAMS — The Regional Atmospheric Modeling System, Version 4.3/4.4, Introduction to RAMS 4.3/4.4*. ASTER Division, Mission Research Corporation, Fort Collins, CO, 2001.

Weschler C.J., Shields H.C., and Nike D.V. Indoor ozone: recent findings. In: R. Burglund (Ed.), *Tropospheric Ozone in the Environment II*. Air and Waste Management Association, Pittsburgh, PA, 1992, pp. 681–700.

Whitfield R.G., Richmond H.M., and Johnson T.R. Overview of ozone human exposure and health risk analyses used in the US EPA's review of the ozone air quality standard. Proceedings of the Fifth US–Dutch International Symposium on Air Pollution in the 21st Century, ANL/DIS/CP-92660, Noordwijk, The Netherlands, 1997.