Characterizing Population Exposures and Co-exposures to Ozone and PM2.5 for the City of Philadelphia Using a Source-to-Dose Framework

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Abstract
This paper provides an overview of a modeling study of population exposure to Ozone ($O_3$) and Particulate Matter 2.5 ($PM_{2.5}$) for the City of Philadelphia and a surrounding area. The study focused on a two-week episode, from July 11, 1999 to July 24, 1999, and employed the consistent source-to-dose modeling framework of MENTOR/OPERAS (Modeling Environment for Total Risk Studies/Ozone and Particles Exposure and Risk Analysis Systems). The MENTOR/OPERAS framework consists of four components needed to estimate population exposure/dose: (1) calculation of ambient outdoor concentrations, (2) spatiotemporal interpolation for obtaining census-tract level outdoor concentrations, (3) calculation of microenvironmental concentrations to match the corresponding activity patterns, and (4) population dosimetry modeling. This study shows the feasibility of developing population exposure assessments to ozone and PM using an integrated and mechanistically consistent approach.

Introduction
Evaluation of human exposure to complex atmospheric contaminants such as primary and secondary air toxics and fine airborne particulate matter (PM) has typically been based on data from fixed ambient monitoring stations. This practice results in artificial characterization of human exposures/doses. The concentrations and physicochemical attributes of photochemical pollutants, air toxics, and aerosol in residential and occupational environments and, furthermore, in the flow that actually enters the human respiratory tract, vary significantly and are in general quite different from corresponding outdoor monitor values.

New data and scientific studies have shed new light on the complexity of interactions between gas phase pollutants such as $O_3$ and PM, and have highlighted the importance of studying the co-occurrence of such contaminants. It is important to note that these interactions also take place at local, microenvironmental, and biological scales that are not characterized well by ambient monitor data. Past studies are limited in their scope because they typically address only one parameter ($O_3$ or $PM_{2.5}$); and because they use data from a limited number of air quality monitors to characterize the local ambient air quality\(^1\). The present study takes advantage of the extensive regional monitoring networks that came online in 1999 for monitoring hourly $PM_{2.5}$ ambient concentrations; data from the intensive North East Oxidant and Particulate Study (NE-OPS) in the Philadelphia region; and regional air...
quality model predictions; to obtain population exposure distributions due to co-occurrence of O₃ and PM₂.₅.

The comprehensive modeling of individual/population exposure to ozone and PM₂.₅ will in general require the ten steps, that are listed below. This list represents a “composite” based on modeling frameworks that appeared in the literature over the last twenty years²,³,⁴,⁵,⁶,⁷.

1. Estimate the background or ambient levels of both ozone and PM₂.₅. This can be done through either:

   (a) geostatistical analysis of fixed monitor data, or
   (b) emissions-based, photochemical, air quality modeling (typically with a regional, grid-based model such as UAM-V, CAMx, MAQSIP, Models-3/CMAQ) applied in a coarse resolution mode.

2. Estimate local outdoor levels of both ozone and PM₂.₅. These levels could typically characterize the ambient air of either an administrative unit (such as a census tract, a municipality, a county, etc.) or a conveniently defined grid cell of an urban scale air quality model. Again, this may involve either

   (a) appropriate (multivariate) geostatistical analysis of monitor data, or
   (b) application of an urban scale model (such as UAM-IV) or a multi-scale, grid based model (such as UAM-V, CAMx, CMAQ) at its highest resolution (typically 2-4 km), or
   (c) “correction” of the estimates of the regional model using some scheme that “adjusts” for subgrid chemistry and mixing processes.

3. Estimate levels and temporal profiles of both ozone and PM₂.₅ in various outdoor and indoor microenvironments such as street canyons, residences, offices, restaurants, vehicles, etc. This can be done through either

   (a) linear regression of available observational data sets,
   (b) simple mass balance models (with linear transformation and sinks) over the volume (or a portion of the volume) of the microenvironment,
   (c) detailed (nonlinear) gas or gas/aerosol chemistry models, or
   (d) detailed combined chemistry and computational fluid dynamics models.

4. Characterize relevant attributes of individuals or populations under study (age, gender, weight, occupation, other physiological characteristics). For Population Based Modeling (PBM) one can either:
(a) select a fixed-size sample population in a way that it statistically reproduces essential demographics (age, gender, race, occupation, income, education) of the population unit used in the assessment (e.g. a sample of 500 people is typically used to represent the demographics of a given census tract), or

(b) divide the population-of interest into an exhaustive set of cohorts representing subpopulations.

5. Develop activity event (or exposure event) sequences for each member of the sample population or for each cohort for the exposure period. This could utilize:

(a) existing databases based on composites of questionnaire information from past studies, or

(b) study-specific information, if available.

6. Calculate appropriate inhalation rates for the individuals of concern, or the members of the sample population, reflecting/combining the physiological attributes of the study subjects and the activities pursued during the individual exposure events.

7. Combine intake rates and microenvironmental concentrations for each activity event to assess exposures.

8. Average exposure estimates over time-units (e.g. 1-hour average, 8-hour running average, etc.) characterizing the exposure metric of concern.

9. Develop appropriate estimates of dose corresponding to calculated exposure and intake estimates, in conjunction with physiological and activity estimates.

10. For Population Based Modeling, extrapolate population sample (or cohort) exposures and doses to the entire populations of interest and quantify, to the extent possible, variability and uncertainty in the various components, assessing their effect on the estimates of exposure and dose.

Implementation of the above components of comprehensive exposure modeling has benefited significantly from recent advances and expanded availability of computational technologies such as Relational Database Management Systems (RDBMS) and Geographic Information Systems. The current study attempt to address/incorporate all the above components in the MENTOR/OPERAS framework.

Methods
The following describe how the ten steps of comprehensive population exposure modeling are implemented under the MENTOR/OPERAS framework in the current study.

Calculation of Ambient Outdoor Concentrations (step 1)
In this study, ambient outdoor concentrations were obtained in two different ways - from regional photochemical model predictions, and from measured ambient concentrations from national networks as well as local studies.

The state-of-the-art Community Multi-scale Air Quality (CMAQ) model was used to obtain predictions for the time period of interest. The 1998 version of the National Emissions Inventory maintained by USEPA was used for the area, mobile and point source terms. The emissions were processed using the Sparse Matrix Kernel Estimator (SMOKE) preprocessor. Meteorological inputs were obtained from the National Climatic Data Center (NCDC) archives, and processed through the National Center for Atmospheric Research (NCAR) MM5 Version 3 meteorological model. The MM5 model utilized as input gridded ECMWF data at 2.5 degree resolution as well as rawinsonde and surface observations. The model utilized the high resolution Blackadar scheme for PBL, Grell’s scheme for cumulus parameterization, mixed phase (Reisner scheme) for explicit moisture and a cloud radiation scheme. The output frequency for MM5 and CMAQ models was one hour. The models utilized a one-way nested grid comprising three grids of 36 km, 12 km and 4 km resolution and had fourteen sigma layers in the vertical. The 4 km domain encompassed the Philadelphia and New Jersey regions while the 36 km domain encompassed Ozone Transport Assessment Group (OTAG) domain. Further details of the regional photochemical modeling study are available in the referenced documents.

The second source for outdoor concentration data for \(O_3\) and \(PM_{2.5}\) was regional and local air quality monitoring networks. 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 air-quality monitoring study conducted by the North-East Oxidant and Particulate Study (NE-OPS) consortium. The \(PM_{2.5}\) hourly monitor data were used primarily for photochemical model evaluation due to the limited number of monitors.

**Spatiotemporal Interpolation for Obtaining Census-Tract Level Outdoor Concentrations (step 2)**

In this study, the regional air quality modeling predictions and observed data were interpolated to the scale of census tracts by using methods of the Spatio-Temporal Random Field (STRF) theory to prepare the input concentrations for the population exposure model. The STRF approach interpolates monitor data in both space and time simultaneously. This method can thus analyze information on “temporal trends”, which cannot be incorporated directly in purely spatial interpolation methods such as standard kriging. Furthermore, the STRF method can optimize the use of data that are not uniformly sampled in either space or time. STRF was further extended in the Bayesian Maximum Entropy (BME) framework and applied to ozone interpolation studies. It should be noted that these studies formulate an over-arching scheme for linking air quality with population dose and health effects; however they are limited by the fact that they do not include any microenvironment effects or linkages to the nonlinear dynamics of ozone exposure. Further details about the theory of spatiotemporal random fields, and the space-time estimation method are available.
in Christakos\textsuperscript{20} and Vyas\textsuperscript{21}.

**Population Exposure Modeling (step 3 - 5)**

Existing comprehensive inhalation exposure models treat human activity patterns as sequences of exposure events in which each event is defined by a geographic location and a microenvironment. USEPA offices (OAQPS and NERL) have supported the most comprehensive efforts in this area, that have resulted in the NEM/pNEM (National Exposure Model and Probabilistic National Exposure Model), HAPEM (Hazardous Air Pollutant Exposure Model) and SHEDS (Stochastic Human Exposure and Dose Simulation model) families of models. Recently SHEDS\textsuperscript{1} has been modified and incorporated into MENTOR-OPERAS. This variant of SHEDS includes both ozone and PM-relevant microenvironmental processes while providing interactive linking with the Consolidated Human Activity Database (CHAD) for consistent definition of population characteristics and activity events\textsuperscript{22,23,24,25}. The CHAD database has been developed for EPA’s National Exposure Research Laboratory by ManTech Environmental Technologies. It contains 22,968 person days of activity. All ages and both genders are included in the database, and information regarding every activity undertaken during a day, and lasting for a minute or more, is included in sequential order.

The interpolated outdoor concentrations of $O_3$ and $PM_{2.5}$ at census-tract level are used as inputs to the MENTOR/SHEDS module for estimating different microenvironmental concentrations that each member of the population has encountered during the sequence of activity events. The MENTOR/SHEDS component of MENTOR/OPERAS consists of two components that estimate $O_3$ and $PM_{2.5}$ levels in various microenvironments. The estimation of $O_3$ concentrations in various microenvironments are based on the general mass balance equation used in pNEM/O$_3$ model as below:

\[
\frac{dC_{in}}{dt} = (F_p)(v)(C_{out}) + \frac{S}{V} - (v + F_d)(C_{in})
\]

where $C_{in}$ = indoor concentration (mass/volume), $F_p$ = penetration factor (dimensionless fraction), $v$ = air exchange rate (1/time), $C_{out}$ = outdoor concentration (mass/volume), $S$ = indoor source generation rate (mass/time), $V$ = indoor volume (volume), and $F_d$ = $O_3$ decay rate (1/time). To simplify this equation, we made the following assumptions: (1) steady-state, (2) S=0 (no indoor sources), and (3) $F_p$=1. Therefore, equation (2) becomes:

\[
\frac{C_{in}}{C_{out}} = \frac{v}{v + F_d}
\]

For different microenvironments, there are different distributions for determining the parameters of $v$ and $F_d$. To be more specific, there are three distributions of the air exchange rate ($v$), corresponding to the residential, non-residential, and vehicle microenvironments. There are two distributions for the $O_3$ decay rates, corresponding to the residential and non-residential including vehicle microenvironments. The values of these two parameters ($v$ and $F_d$) are drawn randomly from their specific 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 as developed by Burke et al. (2001). For the indoor residential microenvironment, a single-compartment, steady-state mass balance equation is used to calculate indoor PM concentrations from the infiltration of ambient PM indoors and indoor PM sources as shown below:

\[ C_{\text{residential}} = \frac{P \times \text{ach}}{\text{ach} + k} C_{\text{ambient}} + \frac{E_{\text{smk}} N_{\text{cig}} + E_{\text{cook}} t_{\text{cook}} + E_{\text{other}} T}{(\text{ach} + k) VT} \] (3)

where \( C_{\text{ambient}} \) = ambient outdoor PM concentration (\( \mu g/m^3 \)), \( P \) = penetration factor (unitless), \( k \) = deposition rate (\( h^{-1} \)), \( \text{ach} \) = air exchange rate (\( h^{-1} \)), \( E_{\text{smk}} \) = emission rate for smoking (\( \mu g/cig \)), \( N_{\text{cig}} \) = number of cigarettes smoked during model time step, \( E_{\text{cook}} \) = emission rate for cooking (\( \mu g/min \)), \( t_{\text{cook}} \) = time spent cooking during model time step (min), \( E_{\text{other}} \) = emission rate for other source (\( \mu g/h \)), \( T \) = model time step (h), and \( V \) = residential volume (\( m^3 \)). The first term in equation (1) 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 \( T \), which is based on the duration of the activity events assigned to the modeled person from CHAD.

For the non-residential 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:

\[ C_{\text{microenvironment}} = b_0 + b_1 * C_{\text{ambient}} \] (4)

**Population Dosimetry Modeling (step 6 - 9)**

The outputs from the MENTOR/SHEDS module provide exposure event sequences for each member of the sample population and the corresponding microenvironmental concentrations of \( O_3 \) and \( PM_{2.5} \) in these event sequences. Exposure to \( O_3 \) and \( PM_{2.5} \) occurs when individuals inhale air containing these constituents. Thus, the amount of pollutant delivered to the lung is dependent upon the person’s inhalation rate. A new lung dosimetry model was developed in the present study to calculate the inhalation rates for general population covering different ages and gender based on the concepts in HUMTRN\(^{26}\), a subroutine of the BIOTRAN model developed by Los Alamos National Laboratories. Then, the calculated inhalation rate is combined with the associated microenvironmental concentrations to estimate the potential dose delivered to the lung for each member of the sample population.

The process of calculating inhalation rates is described in the following procedures.

1. Calculate ideal body mass (\( HWT \)), in units of kg, based on the inputs of age and gender for each member of the sample population:

\[ HWT = \exp(Z_{\text{score}} \times \log(a) + \log(b)) \] (5)

where
• $Z_{\text{score}}$ is the random factor generated from the standard Normal distribution with mean=0 and standard deviation=1,
• $a, b$ are age and gender specific regression coefficients

2. Use the body mass to calculate a daily basal metabolic rate ($DBMR$), in units of mJoule/day, based on the following age-specific empirical equation

$$DBMR = c \times HWT + d + Z_{\text{score}} \times e$$  

(6)

where

• $HWT$ = the body mass,
• $Z_{\text{score}}$ is a random factor generated from the standard Normal distribution with mean=0 and standard deviation=1,
• $c, d, e$ are age and gender specific regression coefficients

3. Convert the daily basal metabolic rate ($DBMR$) to the basal metabolic rate ($BMR$) in units of kcal/min:

$$BMR = DBMR \times 239(\text{kcal/mJoule})/1440(\text{min/day})$$

4. Calculate the conversion factor ($EETOVO_2$) between energy expenditure ($EE$) and oxygen uptake rate ($VO_2$):

for males:

$$EETOVO_2 = 0.2 + U_{\text{score}} \times (0.22 - 0.20)$$

for females:

$$EETOVO_2 = 0.19 + U_{\text{score}} \times (0.21 - 0.19)$$

where $U_{\text{score}}$ is a random factor generated from the Uniform distribution (0,1).

5. Calculate the ventilation rate conversion factor ($VQ$) between air uptake rate (L-air) and oxygen uptake rate (L-O$_2$) 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 in the website: www.epa.gov/chadnet1).

$$\begin{align*}
\text{METs} \leq 4.5 & : \quad VQ = 25 + U_{\text{score}} \times 5 \\
\text{METs} > 4.5 & : \quad VQ = 31 + U_{\text{score}} \times 4
\end{align*}$$
6. The inhalation rate \( (V_E\text{ in unit of L/min}) \) is then calculated based on the following equation:

\[
V_E = BMR \times METS \times EETOVO_2 \times VQ
\]

For calculating the potential 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 and its updated ICRP 1972 database to calculate the PM\(_{2.5}\) amount deposited in the three regions of lung. The potential dose of PM\(_{2.5}\) is then the sum of 3 deposited PM\(_{2.5}\) mass quantities. The potential dose of O\(_3\) is the total amount of O\(_3\) entering the lung (concentration\(\times\)inhalation rate), since O\(_3\) is very reactive.

**Results and Discussion**

The 24-hour aggregated outdoor, indoor, and total doses for O\(_3\) and PM\(_{2.5}\) were obtained. For the O\(_3\) exposure case (see Figure 1), total dose is due to outdoor sources, indicating that there is no contribution from indoor sources to the total O\(_3\) exposure. As shown in Figure 2, the distribution curve of total PM dose keeps apart from that of dose due to outdoor sources, indicating that indoor sources also contribute significantly to the total population dose.

To facilitate comparison of patterns in O\(_3\) and PM\(_{2.5}\) occurrences, the modeled concentrations were first normalized by the standards for both parameters - the 80 ppb level for the maximum daily 8-hour running average for O\(_3\); and the 65 \(\mu g/m^3\) daily average for PM\(_{2.5}\). So, the O\(_3\) concentrations are normalized as \(n_{o3} = (a_{o3} - 80)/80\), where \(a_{o3}\) is the daily maximum value of 8-hour running averages of observed O\(_3\) concentrations. The PM\(_{2.5}\) concentrations are normalized as \(n_{pm25} = (a_{pm25} - 65)/65\), where \(a_{pm25}\) is the daily (24-hour) averaged PM\(_{2.5}\) concentration. The ratios of normalized values were then computed for each census tract, for each day, as:

\[
rn = \frac{n_{o3}}{n_{pm25}}
\]  
(7)

The maps of these ratios provide a means of examining the spatial distribution of O\(_3\) and PM\(_{2.5}\) 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 condition is satisfied, then \(n_{pm25}\) will always be negative and positive values of \(n_{o3}\) indicate locations and magnitude of O\(_3\) exceedances. The ratio \(rn\) 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 results of co-occurrence analysis are presented for 18 July 1999 (see Figure 3). It can be shown that the ratio \(rn\) are all negative across the city of Philadelphia, since O\(_3\) exceeds the standard but PM\(_{2.5}\) does not.
Figure 1: Cumulative distribution function of O$_3$ total doses and total doses from outdoor sources; across urban Philadelphia, 11 July 1999 to 24 July 1999. 500 people were simulated for each of the 482 census tracts used in the study. CMAQ predictions interpolated to census tracts were used as outdoor concentrations.
Figure 2: Cumulative distribution function of PM$_{2.5}$ total doses and total doses from outdoor sources; across urban Philadelphia, 11 July 1999 to 24 July 1999. 500 people were simulated for each of the 482 census tracts used in the study. CMAQ predictions interpolated to census tracts were used as outdoor concentrations.
Figure 3: Ratio of normalized $\text{O}_3$ concentrations to normalized $\text{PM}_{2.5}$ concentrations, for 18 July 1999, over Urban Philadelphia.
A second metric for characterizing co-occurrence patterns is provided through the use of empirical spatiotemporal cross-covariances, which are computed as follows:

\[
C(h, \tau) = \frac{1}{N_{h,\tau}} \sum_{i=1}^{N_{h,\tau}} \left\{ X(s_i, t_i) - \bar{X} \right\} \left\{ Y(s_j, t_j) - \bar{Y} \right\}
\]

(8)

where \( \bar{X} = \sum_{i=1}^{n} X(s_i, t_i) \), \( \bar{Y} = \sum_{j=1}^{n} Y(s_j, t_j) \), \( h = s_i - s_j \), and \( \tau = t_i - t_j \). The spatiotemporal cross covariances provide a measure of the strength of correlation between O\(_3\) and PM\(_{2.5}\) concentrations and total doses. The results (see Figure 4) indicate that there is loss of correlation during evening/night, when ozone is titrated by NOx.

**Conclusions**

This study showed the feasibility of developing population exposure assessments to ozone and PM using an integrated and mechanistically consistent source-to-dose framework, provided by MENTOR-OPERAS. The results show the necessity to incorporate extensive additional information to that derived from ambient monitor observations when studying the exposures and in elucidating the causes of associated health impacts on populations due to air pollutants such as ozone and PM.

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Figure 4: Cross-covariance between normalized daily maximum values of 8-hour running averages of O$_3$ concentrations and daily maximum values of PM$_{2.5}$ 1-hour averaged concentrations; for 11 July 1999 to 23 July 1999; across urban Philadelphia
References

**Key Words**
Population Exposure Assessment/Modeling, Photochemical Air Quality Modeling, O$_3$ Exposure Models, PM$_{2.5}$ Exposure Models.