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Modeling population exposures to outdoor sources of hazardous air pollutants

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Accurate assessment of human exposures is an important part of environmental health effects research. However, most air pollution epidemiology studies rely upon imperfect surrogates of personal exposures, such as information based on available central-site outdoor concentration monitoring or modeling data. In this paper, we examine the limitations of using outdoor concentration predictions instead of modeled personal exposures for over 30 gaseous and particulate hazardous air pollutants (HAPs) in the US. The analysis uses the results from an air quality dispersion model (the ASPEN or Assessment System for Population Exposure Nationwide model) and an inhalation exposure model (the HAPEM or Hazardous Air Pollutant Exposure Model, Version 5), applied by the US. Environmental protection Agency during the 1999 National Air Toxic Assessment (NATA) in the US. Our results show that the total predicted chronic exposure concentrations of outdoor HAPs from all sources are lower than the modeled ambient concentrations by about 20% on average for most gaseous HAPs and by about 60% on average for most particulate HAPs (mainly, due to the exclusion of indoor sources from our modeling analysis and lower infiltration of particles indoors). On the other hand, the HAPEM/ASPEN concentration ratio averages for onroad mobile source exposures were found to be greater than 1 (around 1.20) for most mobile-source related HAPs (e.g. 1, 3-butadiene, acetaldehyde, benzene, formaldehyde) reflecting the importance of near-roadway and commuting environments on personal exposures to HAPs. The distribution of the ratios of personal to ambient concentrations was found to be skewed for a number of the VOCs and reactive HAPs associated with major source emissions, indicating the importance of personal mobility factors. We conclude that the increase in personal exposures from the corresponding predicted ambient levels tends to occur near locations where there are either major emission sources of HAPs or when individuals are exposed to either on- or nonroad sources of HAPs during their daily activities. These findings underscore the importance of applying exposure-modeling methods, which incorporate information on time–activity, commuting, and exposure factors data, for the purposes of assigning exposures in air pollution health studies.
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Introduction

Estimating human exposure is a critical component of most environmental risk assessments. Human exposures to hazardous air pollutants (HAPs) can result from contact with air, water, soils, and food. Exposures may be dominated by contact with a single medium (e.g., ambient air) or result from concurrent contacts with multiple media (e.g., air, water, soil). The nature and extent of such exposures depend largely on two things: human factors and the concentration of a pollutant in the relevant exposure media. Human factors include behavioral, sociological, and physiological characteristics of individuals or groups of people that directly or indirectly affect their contact with the substances of concern.

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The concentration of a pollutant in an exposure medium can be modeled or measured at various locations, or microenvironments (MEs) (e.g., outdoors near home, commuting, indoors at home, indoors at work, school), in which the individuals are expected to spend an appreciable amount of their time. This paper focuses on jointly examining the results from an air quality dispersion model, the ASPEN or Assessment System for Population Exposure Nationwide model (Rosenbaum et al., 1999; US EPA, 2000) and an inhalation exposure model, the Hazardous Air Pollutant Exposure Model, Version 5 (HAPEM), applied during the 1999 National Air Toxic Assessment (NATA) study (US EPA 2006a) for assessing exposures and risks from many particulate and gaseous HAPs in the US. The goal of our analysis is to examine the significance of the differences between modeled exposures to outdoor HAPs and the corresponding ambient pollutant concentrations that is relevant to air pollution epidemiology research. Specifically, we examine the role of human mobility, such as commuting to work, and exposure-related factors, such as infiltration and microenvironmental factors, in either increasing or

decreasing individuals' exposures to concentrations of outdoor HAPs at their home census tract. Both NATA and the analysis reported here focus only on the contribution of outdoor sources of HAPs to modeled personal exposures. In this paper, we evaluate the relationship between modeled outdoor concentrations and personal exposures of HAPs using the ASPEN and HAPEM models. Figure 1 describes the analysis methodology schematically.

Methods

Air Quality Modeling

Before performing air quality modeling as part of NATA, the emission inventories are processed in Emissions Modeling System for Hazardous Air Pollutants (EMS-HAP; US EPA, 2004a) to create the emissions input files. EMS-HAP spatially allocates area and mobile source emissions inventoried at the county level to the census tract level using surrogate data, such as population, industrial land use, and roadway miles, and temporally allocates them by using temporal profiles that are matched to the emissions data by codes, to eight 3 h time periods throughout the day to account for diurnal variability in emissions. EMS-HAP also groups source categories into source sectors, such as ''onroad mobile'', ''nonroad mobile'', ''major'', and ''area and other'' sources. The ''area and other'' source sector combines ''area'' and ''other'' sources. Area sources are smaller stationary sources that emit less than 10 tons per year of a single air pollutant or less than 25 tons per year of a combination of HAPs. Examples of area sources include neighborhood dry cleaners and gas stations.

Once the emissions are processed, they are input into the ASPEN model (Rosenbaum et al., 1999; US EPA, 2000). The ASPEN model takes into account important determinants of pollutant concentrations, such as the rate of release,

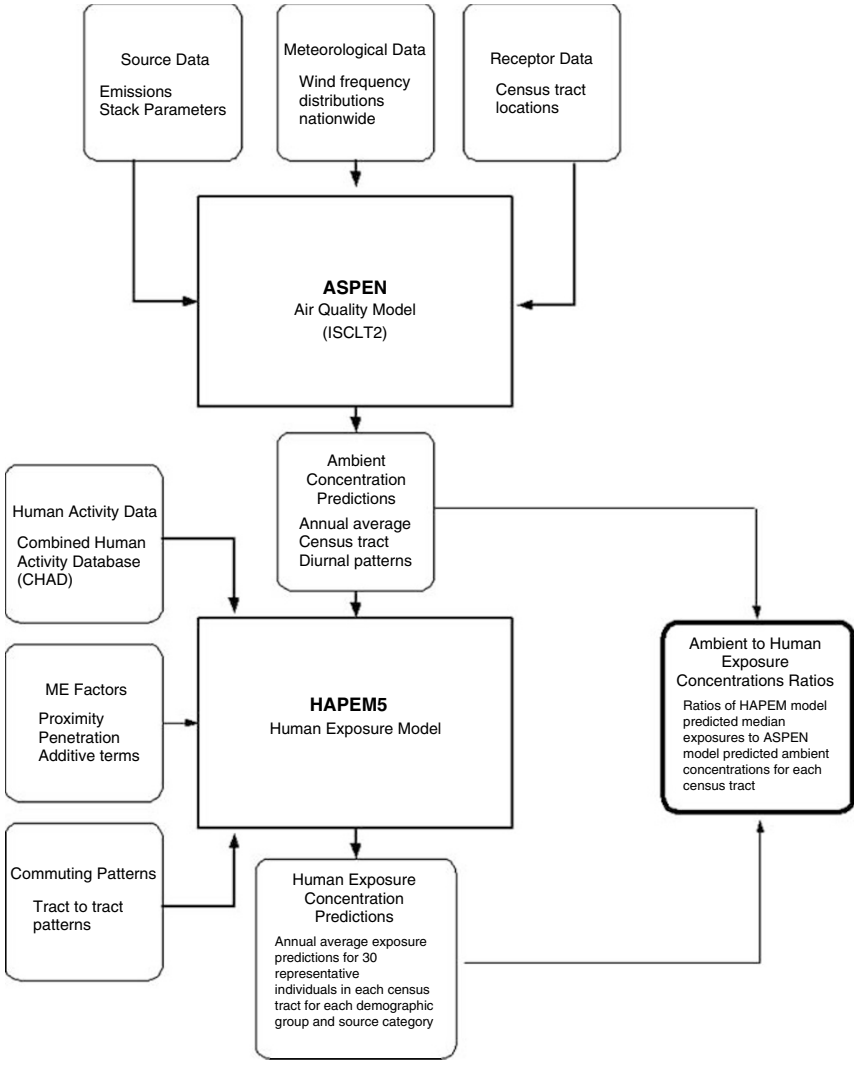

Figure 1. Components of air quality and exposure modeling tools used to calculate model predicted exposure to concentration ratios.

location of release, the height from which the pollutants are released, meteorological variables such as wind speeds, wind direction, and atmospheric stability from the meteorological stations nearest to the release, breakdown of the pollutants in the atmosphere after being released (i.e., reactive decay), settling of pollutants out of the atmosphere (i.e., deposition), and transformation of one pollutant into another (i.e., secondary formation). ASPEN calculates annual average concentrations at census tract internal point, or centroid, locations. Meteorological conditions in 1999 are used to match the emission period, and census tract data from 2000 are used. Meteorological data for 1999 were used to generate frequency distributions stratified by time of day into eight 3 h time blocks. This along with similar emission rate stratification helps preserve any characteristic diurnal patterns that might be important in subsequent estimation of population exposure. The resulting output of ASPEN is a grid of annual average concentration estimates for each source/pollutant combination by diurnal time block (US EPA, 2000).

ASPEN only accounts for sources within a 50 km radius of each source when calculating ambient concentrations. Thus, the contribution to ambient levels of HAPs from sources further away than 50 km, as well as the contribution of uninventoried sources is addressed through the addition of a ''background'' term (Battelle, 2003). An empirical approach (Rosenbaum et al., 1999) is used to estimate the secondary contribution through photochemical transformation of reactive pollutants such as acetaldehyde, acrolein, formaldehyde, and propionaldehyde. Model to monitor comparisons showed that the model performed within a factor of two for inert gases such as benzene, but underpredicted for metals.

Exposure Modeling

HAPEM is a screening-level exposure model appropriate for assessing average long-term inhalation exposures of the general population, or a specific subpopulation, over spatial scales ranging from local to national. HAPEM uses the general approach of tracking representatives of specified demographic groups as they move among indoor and outdoor MEs (i.e., a location in which human contact with an ambient pollutant may take place). The estimated pollutant concentrations in each ME visited are combined with the fraction of time spent in that ME to calculate a time-weighted average exposure concentration for a representative individual assigned to a particular demographic group (US EPA, 2004b).

HAPEM uses four primary sources of information: population data from the US Census, population activity data, air quality data, and microenvironmental data.

Population Data

The US Census Bureau is the primary source of most population demographic data, which include information on where people live, their demographic makeup (e.g., age, gender, ethnic group), and employment. The default population data for HAPEM uses 2000 US Census data reported at the spatial resolution of census tracts, which are small, relatively permanent statistical subdivisions of a county. Census tracts usually contain between 2500 and 8000 residents.

Activity Data

HAPEM uses two types of population activity data: activity-pattern data and commuting-pattern data. Human activity-pattern data are used to determine the frequency and duration of exposure for specific groups within various MEs. Activity-pattern data are taken from demographic surveys of individuals' daily activities, the amount of time spent engaged in those activities, and the locations where the activities occur.

In addition to recording the duration and location of a person's activities, these surveys also collect important demographic information about the person. The demographic information usually includes the person's age, gender, and ethnic group. Most activity-pattern studies also try to collect information on other attributes of a respondent, such as highest level of education completed, number of people in their household, whether the person or anyone in their household is a smoker, employment status, and the number of hours spent outdoors.

The default population activity file for HAPEM is derived from a database of activity-pattern surveys called the Consolidated Human Activity Database (CHAD) (Glen et al., 1997). The CHAD is currently comprised of over 22 000 person-days of activity-pattern data, including 140 activities and 114 locations, collected and organized from 12 human activity-pattern surveys. The CHAD contains the sequential patterns of activities for each individual, and each activity has a corresponding location code so that the ME of each activity is known. Because available activity data are not adequate to estimate the exposure of each individual in a population, HAPEM groups activity-patterns data together for people with similar demographic characteristics that are expected to influence exposure to air pollutants (e.g., age, gender, work status), and provides separate exposure estimates for these groups. The activity profiles for each person in a demographic group have an equal chance of being selected from the activity database. The result is that HAPEM generates a distribution of exposure concentrations for each demographic group in each census tract.

HAPEM divides the population into 10 demographic groups (cohorts), based on combinations of age (five categories) and gender. Activity-pattern data are also separated into 3-day types: summer weekdays, other weekdays, and weekends. The commuting data contained in the HAPEM default file was derived from a special 1990 US Census study that specifies the number of residents of each tract that work in that tract and every other US Census tract,

that is, the population associated with each home tract/work tract pair. HAPEM uses these data in coordination with the activity-pattern data to place an individual either in the home tract or the work tract at each time step.

Air Quality Data

The HAPEM requires annual-averaged, diurnally distributed air quality data. In addition, HAPEM can also evaluate the contributions of subsets of the air quality data (e.g., air concentration values for specific source sectors such as point source, area source, mobile source). Although the air concentration data for HAPEM must be in a specific format (e.g., annual average and diurnally distributed), the source of the data could be either from an air dispersion model or an ambient monitor. In the 1999 NATA application utilized here, the annual average ambient air concentrations estimated for each US census tract by ASPEN was used in HAPEM.

To preserve any characteristic diurnal patterns in ambient concentrations that might be important in the estimation of population exposure, ASPEN annual average concentration estimates are stratified by time of day, with an annual average for each of 3-h time blocks (e.g., midnight to 3 am, 3 am to 6 am, etc.). ASPEN air quality files are also provided for each of the four source categories (i.e., major, area, mobile onroad, mobile nonroad). Thus, the results of HAPEM can be summarized for each of the four categories or a combination of them.

Microenvironmental Data

MEs are generally described as a small space in which human contact with a pollutant takes place, and which can be treated as a well-characterized, relatively homogenous locations with respect to pollutant concentrations for a specified time period. The 37 MEs locations used in the national-scale assessment are listed in Table 1, and include indoors at home, school, work, inside an automobile or bus, outdoors, etc.

To calculate a person's exposure concentration, an estimate is required of the concentration in each of the various MEs (US EPA 2006b). In HAPEM, the concentration in each ME is derived from the ambient concentration estimate for the census tract (predicted by ASPEN) using a set of three adjustment factors: PEN, PROX, and ADD.

$$C_{(i,k,t)} = [\mathrm{ASPEN}]_{i,t} \times \mathrm{PEN} \times \mathrm{PROX} + [\mathrm{ADD}]$$

Where:

$C(i,k,t)$ = concentration predicted within census tract i and ME k in time step t ($\mu g/m^3$)

$[ASPEN]i,t$ = ambient concentration estimated from ASPEN in census tract i for time step t ($\mu g/m^3$)

PEN = penetration factor

PROX = proximity factor

ADD = additive factor accounting for sources within the ME ($\mu g/m^3$)

Table 1. HAPEM microenvironments.

Microenvironment no.	Microenvironment	
	Specific	General
1	Car	In vehicle
2	Bus	In vehicle
3	Truck	In vehicle
4	Other	In vehicle
5	Public garage	Indoors
6	Parking lot/garage	Outdoors
7	Near road	Outdoors
8	Motorcycle	Outdoors
9	Service station	Indoors
10	Service station	Outdoors
11	Residential garage	Indoors
12	Other repair shop	Indoors
13	Residence – no gas stove	Indoors
14	Residence – gas stove	Indoors
15	Residence – attached garage	Indoors
16	Residential – stove and garage	Indoors
17	Office	Indoors
18	Store	Indoors
19	Restaurant	Indoors
20	Manufacturing facility	Indoors
21	School	Indoors
22	Church	Indoors
23	Shopping mall	Indoors
24	Auditorium	Indoors
25	Health care facility	Indoors
26	Other public building	Indoors
27	Other location	Indoors
28	Not specified	Indoors
29	Construction site	Outdoors
30	Residential grounds	Outdoors
31	School grounds	Outdoors
32	Sports arena	Outdoors
33	Park/golf course	Outdoors
34	Other location	Outdoors
35	Not specified	Outdoors
36	Train/subway	In vehicle
37	Airplane	In vehicle

The penetration factor, PEN, is an estimate of the ratio of ME concentration to the concurrent outdoor concentration in the immediate vicinity of the ME. Pollutant-specific penetration factors are derived from available results from field studies (US EPA, 2004a). The proximity factor, PROX, is an estimate of the ratio of the outdoor concentration in the immediate vicinity of the ME to the outdoor concentration represented by the air concentration data at a central location (e.g., at a fixed monitoring station site or a grid centroid). In this application, the ASPEN model was used to predict concentrations at the census tract centroid. To predict the ambient pollution levels found outside of a vehicle (on a

roadway), a proximity factor greater than 1 is applied to the tract-level ASPEN predictions, to represent the concentration on a roadway. The roadway ME is influenced by mobile emissions more than other nonroad locations in the census tract. ADD is an additive factor that accounts for emission sources within or near a particular ME, that is, indoor emission sources. For the national scale assessment, which focused primarily on impacts of outdoor sources alone, the ADD term has been set to zero. However, as additional indoor source data become available, this factor can be included in future assessments. PROX and PEN factors have been compiled for 133 HAPs and are summarized in US EPA (2004b). Unfortunately, the data to support quantitative ME factors are not well developed for many of the HAP compounds and for most of the 37 MEs. Thus, representative values are used in many cases based on measured values of similar compounds in similar types of locations. Further, for many of the compounds and MEs, few measurements are available and the representativeness of the available measurements is unknown. Consequently, these database limitations may introduce varying types and degrees of uncertainties into this exposure analysis.

Stochastic Elements

Although it would be difficult to accurately represent the activities of a specific individual due to day-to-day variation, the general behavior of population groups can be well represented using stochastic processes to account for intra- and intersubject variations. This allows the estimates of population exposures to be characterized as distributions rather than point estimates. HAPEM incorporates several stochastic elements as discussed below.

Activity Data One of the stochastic elements is the selection of activity patterns to represent each demographic group. HAPEM estimates long-term average concentrations, but most of the available population activity data sequences are for 24-h periods only. The general approach used by HAPEM is comprised of several steps. The first is to select three sets of 24-h activity patterns, where each set is used to construct an average pattern for an individual for one of three specified day types: weekends, summer weekdays, non-summer weekdays. A set of patterns, rather than a single pattern, is selected for each day type to reflect the day-to-day variability of activity patterns for an individual. The set of patterns is then combined into an average pattern for the day type (US EPA, 2004b). Next, the corresponding exposure concentration is calculated for each of the three day-type average activity patterns. Then, a weighted average of the three exposure concentrations is calculated to represent the annual average concentration, where the weightings represent the number of days per year for each day type (i.e., 104 for weekends, 65 for summer weekdays, 196 for non-summer weekdays). This process is repeated to create a set of 30 annual exposure concentration estimates for each demographic group in each census tract. The value of 30 was selected to provide a statistically robust representation of the potential distribution of human activity in each census tract.

Another stochastic process is applied in HAPEM for demographic groups whose activity patterns indicate time spent at work. For those groups, a work tract is randomly selected for each home tract, using the proportion of workers commuting to each work tract based on the US 2000 census information.

Air Quality Data HAPEM has the ability to characterize multiple air quality concentrations at a census tract. The data for each tract are entered as a data set with up to 500 sets of values for each tract (i.e., sets of eight annual average 3-h time blocks). For each demographic group/replicate, a different set of air quality concentrations is selected for the tract to reflect the variability in air quality among residential locations within the tract. For NATA application, only a single air quality concentration (e.g., annual average) at the census tract centroid was considered.

Microenvironment Factors HAPEM has the capability of representing the distribution of PEN or PROX values for a particular ME-HAP group as either: (1) an empirical distribution (i.e., individual values) given that the number of values does not exceed indoor to outdoor ratios of 10 (due to limitations in the current model program and input file structure); or (2) one of four statistical distributions (normal, lognormal, uniform, triangular). The ME and proximity factors used in this modeling analysis were based on numerous field studies (Palma, 2004). For each tract/demographic group/replicate, and in some cases source type, a different set of ME factors was randomly selected.

Results

We compare the median (i.e., the 50th percentile value of the 30 exposures) HAPEM exposures at each tract with the predicted annual average ASPEN value for that tract. Table 2 shows the national HAPEM to ASPEN concentration ratio averages for each of the study pollutants. This ratio is presented for total sources of emissions, as well as for each of the five emission source sectors considered in the assessment (i.e., major, area and other, onroad mobile, nonroad mobile, background). In general, the HAPEM exposure predictions are lower than the corresponding predicted ASPEN air quality estimates. Note that these findings differ from the results of other published total personal exposure measurement studies (e.g., Wallace et al., 1985; US EPA 1987; Wallace, 1989; Clayton et al., 1999; Özkaynak, 1999; Kinney et al., 2002), as our modeling-based estimates did not include

Table 2. Ratio of national average HAPEM concentration to national average ASPEN concentration for each source category and across all source categories.

Pollutant	Major	Area and other	Onroad	Nonroad	Estimated background	Total
Gases						
1,1,2,2-Tetrachloroethane	0.87	0.86			0.78	0.78
1,3-Butadiene	0.82	0.84	1.24	0.95	0.75	0.97
1,3-Dichloropropene	0.82	0.84				0.84
Acetaldehyde	0.85	0.84	1.14	0.87	0.77	0.96
Acrolein	0.85	0.85	1.15	0.87		1.01
Acrylontrile	0.85	0.85				0.85
Benzene	0.84	0.87	1.19	0.91	0.76	0.99
Carbon tetrachloride	0.88	0.89			0.78	0.78
Chloroform	0.86	0.86			0.77	0.82
Ethylene dibromide	0.86	0.92			0.78	0.78
Ethylene dichloride	0.86	0.89			0.78	0.78
Ethylene oxide	0.85	0.85				0.85
Formaldehyde	0.87	0.85	1.24	0.88	0.77	0.93
Hexachlorobenzene	0.85	0.88			0.78	0.85
Hydrazine	0.85	0.80				0.85
Methylene chloride	0.86	0.89			0.77	0.83
Perchloroethylene	0.86	0.87			0.76	0.84
Propylene dichloride	0.87	0.90			0.78	0.78
Quinoline	0.83	0.83				0.83
Trichloroethylene	0.83	0.92			0.75	0.83
Vinyl chloride	0.87	0.89			0.78	0.79
Particulates						
Arsenic	0.41	0.41				0.41
Beryllium	0.41	0.40		0.43		0.41
Cadmium	0.41	0.40		0.45		0.41
Chromium III	0.40	0.41	0.65	0.42		0.42
Chromium VI	0.41	0.41	0.66	0.42		0.43
Diesel PM[a]			0.69	0.45		0.54
Lead	0.41	0.42		0.46		0.43
Manganese	0.40	0.41	0.68	0.47		0.41
Nickel	0.40	0.42	0.66	0.44		0.42
Mixed						
Coke oven emissions	0.61	0.63				0.61
Mercury	0.64	0.64			0.57	0.58
PCBs	0.62	0.63			0.58	0.58
Total POM	0.59	0.63	0.88	0.66		0.64

[a]For Diesel PM, background is included in the onroad and nonroad mobile concentrations, because source-specific backgrounds are calculated.

the contributions from various sources of indoor air toxics (e.g. VOCs, PM). Our results show that most gaseous pollutants have modeled exposure concentrations up to 35% lower than the corresponding predicted ambient concentrations. For particulates, HAPEM predicts exposure concentrations that are 46% to 59% lower than the corresponding ambient concentrations. For Chromium VI, HAPEM predicts exposures levels to be 52% lower than ambient values. This is due to the fact that many particulate pollutants can not readily penetrate into indoor environments (Özkaynak et al., 1996; Meng et al., 2005).

Figures 2 to 7 are the pollutant-specific ratio distributions for major, area and other, onroad mobile, nonroad mobile, background, and total categories. HAPs are split into three groups, gaseous, particle, and mixed. Vertical gray lines delineate the groups. Before calculating the ratios, the concentration data set was slightly trimmed. First, only nonzero tracts were considered, to avoid dividing by zero. Next, for the remaining tracts, the 5th and 95th percentiles of the ASPEN concentrations were calculated. Ratios were then calculated only for tracts greater than the 5th percentile and less than the 95th percentile of the concentrations for each pollutant and source group to facilitate the interpretation of exposure values which differ most from the corresponding outdoor concentrations. This step was also taken to avoid large ratios that may result from model limitations in

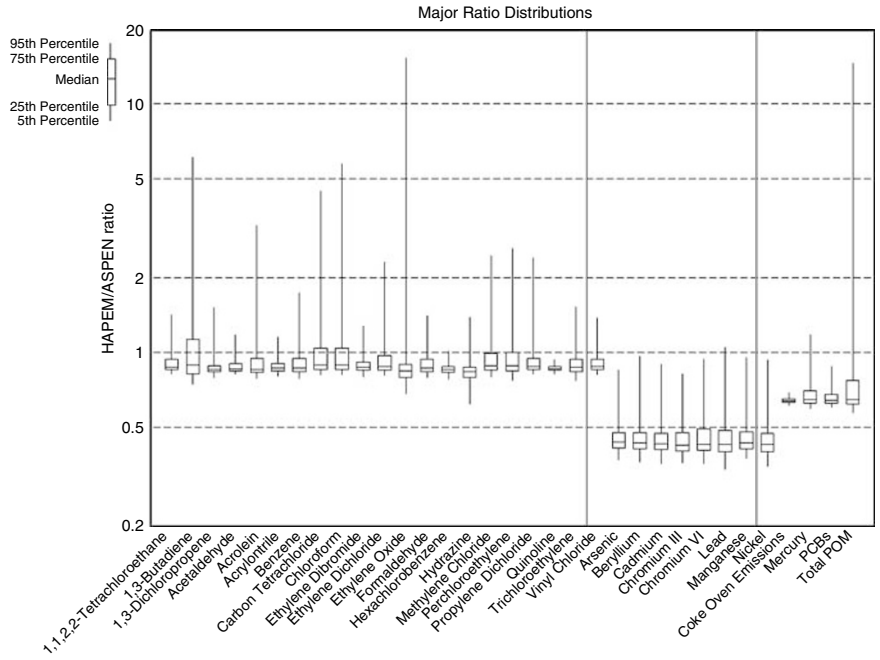

Figure 2. Distributions of HAPEM to ASPEN concentration ratios for major sources.

ASPEN, that is, concentrations only calculated 50 km or less from the tract. However, this step also eliminates tracts solely affected by isolated emission sources situated close to the tract. In Figure 2, the major source ratio distributions are shown from the 5th to the 95th percentiles of the ratio distributions. The ratios for particulate HAPs are generally lower than the gaseous HAPs while mixed HAPs are generally in between. For gaseous HAPs, most of the ratios are below 1, indicating that the predicted exposure concentrations are lower than the modeled ambient concentrations. For almost all HAPs, the range from the 75th to 95th percentile ratios was larger than for the 5th to the 25th percentile. Total POM, ethylene oxide, and 1,3-butadiene are among the pollutants that had the largest difference in the distribution of the upper and lower tails of the ratio distributions. The discrepancy in the magnitude of the skewness in the upper vs. lower tails of the exposure to concentration ratios can be accounted for by the influence of commuting behaviors on personal exposures. Specifically, the extreme values of exposure to concentration ratio are predicted for census tracts that are near major point sources of HAPs emissions. The modeling results indicate that a greater number of individuals who commute to more polluted work districts than to other census tracts with lower pollution than in their home districts.

Area and other source ratio distributions are shown in Figure 3. Again, particulates have lower ratios than gases, with mixed HAPs in between. The distributions are tighter than for major sources, presumably because most area and other source emissions are more homogeneously distributed in adjacent census tracts, so emissions tend to be spread out over a larger area rather than a focal point, such as in the case of major point sources.

Ratios for both onroad and nonroad mobile sources (Figures 4 and 5, respectively) tend to be lower than one for particulate HAPs, but higher (near one or above in the case of onroad mobile sources) for the gaseous HAPs. Background distributions tend to be small (Figure 6). This is likely due to all tracts within a county being assigned the same background concentration for a particular pollutant. So if for a particular home tract, most, if not all, of the work tracts are located in the same county, changes in background concentrations will be low. Total ratios (Figure 7) show less range than major source ratio distributions and fit the same pattern of the area and other, and mobile ratio distributions. The likely reasons for this is that for a lot of tracts, the area and other, mobile, and background concentrations tend to dominate over major source concentrations, so the large differences in the predicted exposure and ambient concentrations for major sources tend to be diluted by the other source categories.

Because the above comparisons are based on an examination of the median HAPEM predictions, they are somewhat limited, because they do not include the full range of possible exposures at a given location. To examine the nature of this problem further, we selected the 90th percentile exposures at a given tract (from the 30 HAPEM predicted exposures at each tract) to determine a change in the distribution of the

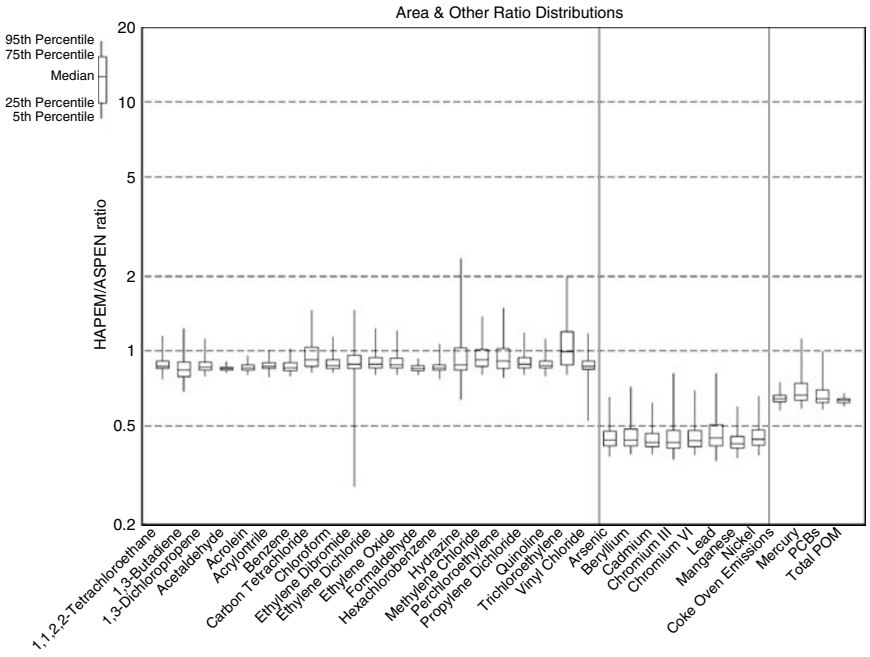

Figure 3. Distributions of HAPEM to ASPEN concentration ratios for area and other sources.

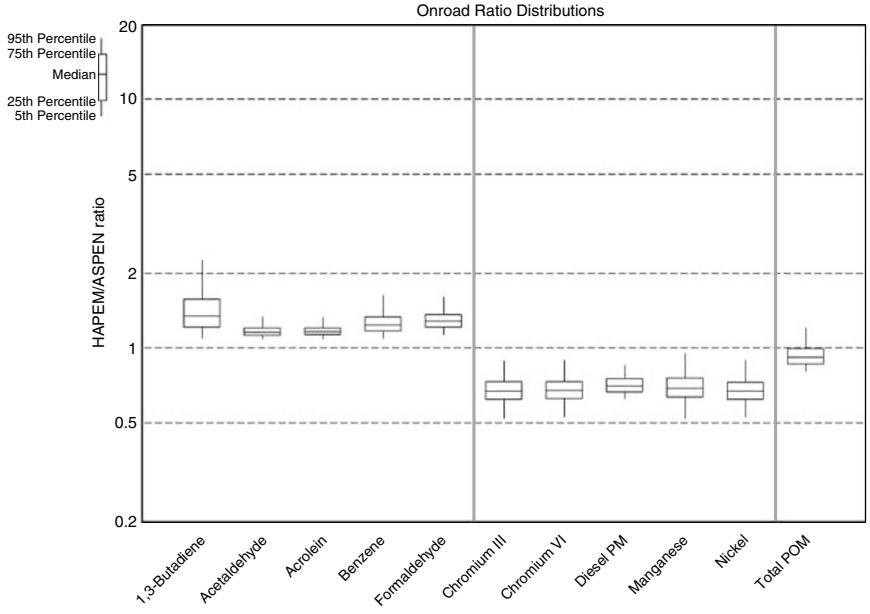

Figure 4. Distributions of HAPEM to ASPEN concentration ratios for onroad mobile sources.

ratios for two selected pollutants, 1,3-butadiene and chromium VI. A comparison of HAPEM/ASPEN ratios for 1,3-Butadine changed from about 1.3 when comparing the median exposure to about 2.4 when examining the 95th percentile exposures at a given tract. Thus, for a pollutant whose penetration and proximity factors are near unity, other factors such as commuting outside of the home tract seem to affect ones' exposure. However, for a particulate metal species, such as hexavalent chromium, the ratio changed from about 0.7 to about 2, indicating that the lower penetration factor for most particulate metals will influence the exposure results. These ratios are depicted in Figure 8.

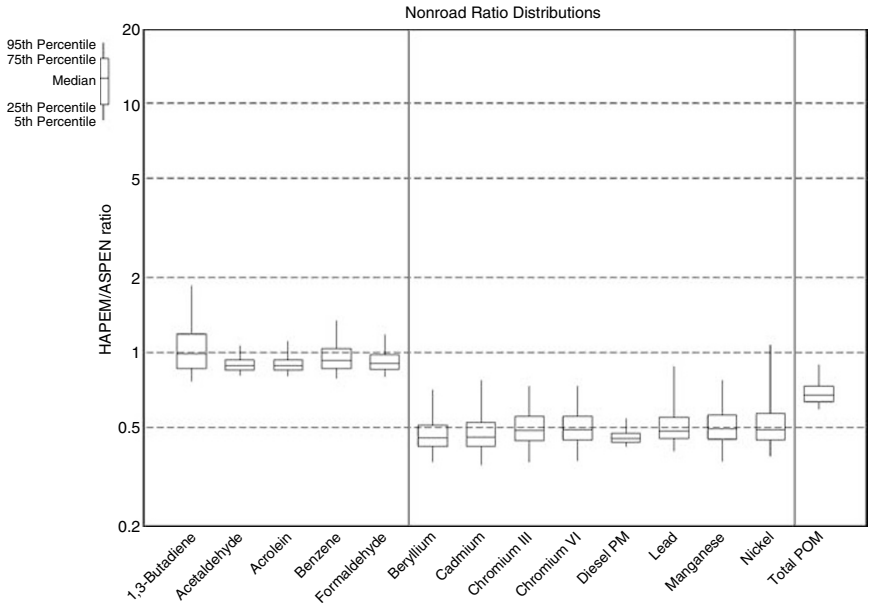

Figure 5. Distributions of HAPEM to ASPEN concentration ratios for nonroad mobile sources.

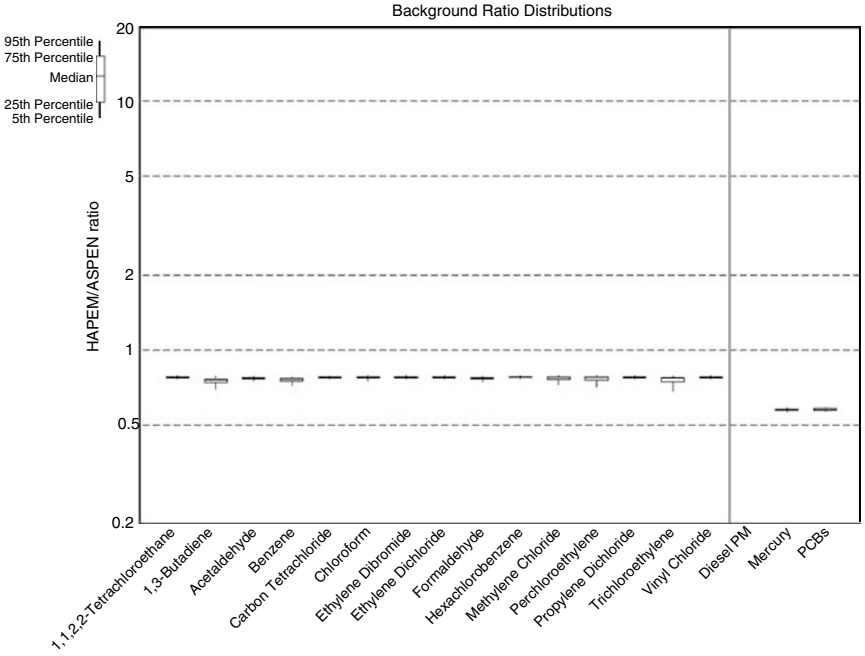

Figure 6. Distributions of HAPEM to ASPEN concentration ratios for background sources.

To further explore the effects of commuting to work on someone's overall exposure concentration, we examined the tract with the highest HAPEM/ASPEN ratio for 1,3-butadiene nationwide. At this chosen tract, the HAPEM concentrations were over 10 times greater than the predicted ASPEN concentration. Figure 9 depicts the distribution of HAPEM/ASPEN concentration ratios for 1,3-butadiene for selected census tracts. The rural tract labeled ''Home'' was shown to have a ratio of 10.6. An examination of the ASPEN predictions at these tracts is shown in Figure 10. A close examination of the home tract, finds a major portion of it commuting population commute to the tract in which the ASPEN concentration was nearly two orders of magnitude higher. This results in relatively high exposure concentrations

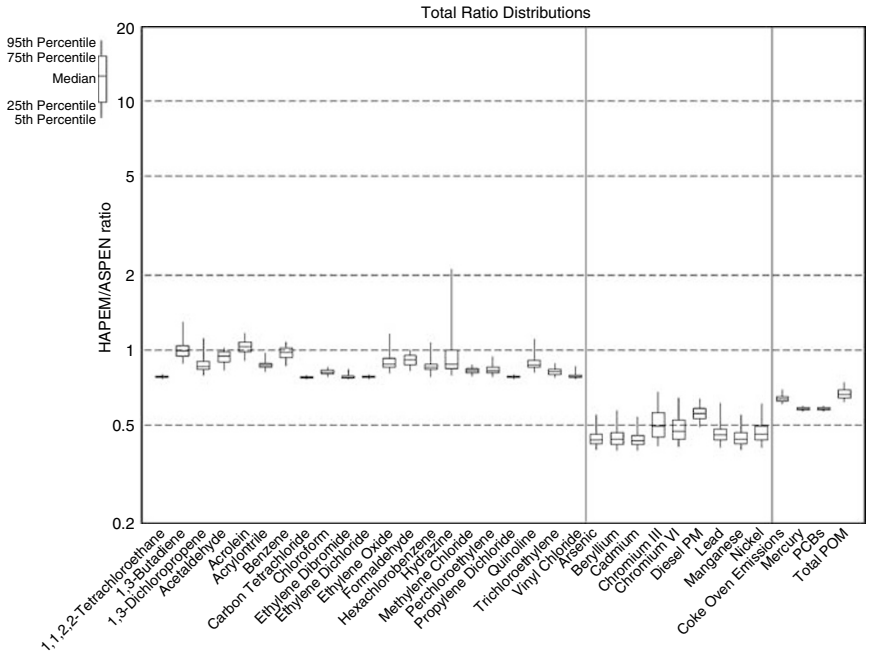

Figure 7. Distributions of HAPEM to ASPEN concentration ratios for all sources (total).

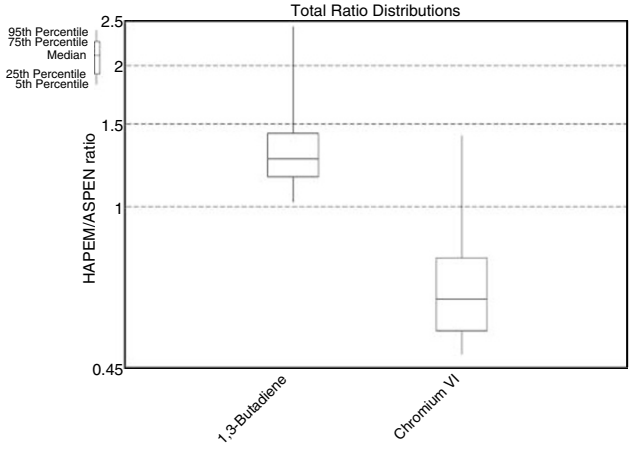

Figure 8. Distributions of HAPEM to ASPEN concentration ratios for all sources (total) (90th percentile exposures).

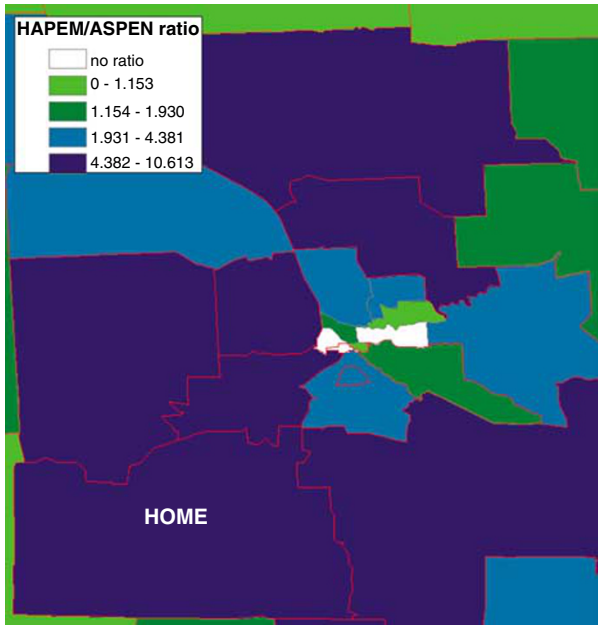

Figure 9. HAPEM to ASPEN concentration ratios for selected census tracts.

for the commuters in the ''Home'' tract resulting in the high HAPEM/ASPEN ratios. Thus, commuting patterns have been found to have a significant impact on one's exposure to outdoor hazardous pollutant concentrations.

Discussion

In this paper, we examined the relationship between modeled personal exposures and outdoor concentrations of a large number of particulate and gaseous HAPs. The analyses employed the results from the 1999 NATA assessment, in which the ASPEN model was used for modeling outdoor pollutant concentrations and the HAPEM model for predicting population exposures to outdoor HAPs. Predicted exposure concentrations of outdoor HAPs from all sources are lower than the modeled ambient concentrations, by about 20% on average for most gaseous HAPs, and by about 60% on average for most particulate HAPs. For the remaining

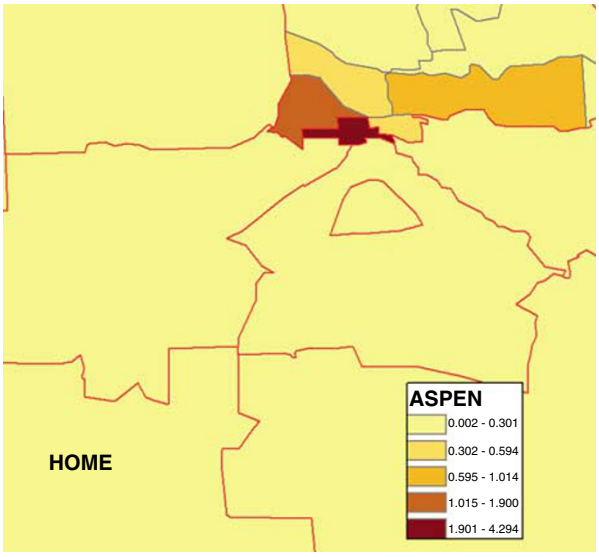

Figure 10. ASPEN concentrations for selected census tracts.

mixed PAHs (e.g., coke oven emissions, mercury, PCBs, total POM) personal exposures on average are predicted to be 40% lower than the corresponding outdoor levels. The lower personal to outdoor ratios obtained for particulates are due to the indoor infiltration efficiencies of most particles into buildings are below 0.7 due to lower penetration and deposition on indoor surfaces (Özkaynak et al., 1996; Özkaynak, 1999). However, an examination of the ratios of personal exposures to outdoor concentrations by different categories of emissions type revealed additional insights. In particular, the HAPEM/ASPEN concentration ratio averages were found to be greater than 1 (around 1.20) for most motor-vehicle related HAPs (e.g. 1, 3-butadiene, acetaldehyde, benzene, formaldehyde) reflecting the importance of near-roadway and commuting environments on personal exposures to HAPs. Furthermore, the distribution of the ratios of predicted personal to ambient concentrations were found to be skewed for a number of the VOCs and reactive HAPs typically associated with major sources of emissions, again indicating the importance of personal mobility factors, such as commuting, on time-weighted microenvironmental exposures. In particular, we have found that commuting to work locations, where major sources are located, could elevate personal exposures to many HAPs (e.g., 1, 3-butadiene, carbon tetrachloride, chloroform, ethylene dichloride, ethylene oxide, methylene chloride, perchloroethylene, propylene dichloride, total POMs) to levels above twice the levels modeled at those individuals home locations or census tracts. A closer examination of the predicted personal to outdoor concentration ratios for both the median and 90th percentile exposures indicated that commuting and/or travel to another location which has higher emissions of HAPs than a the home tract could lead to significant change in these ratios. In particular, we have found that the majority of the distribution of personal to outdoor 1,3-butadiene concentration ratios was over 1, while the upper quartile of these ratios was above 1.4 for the 90th percentile of exposures. We then identified the tract with the highest personal to outdoor 1,3-butadiene concentration ratio from our nationwide modeling analysis. At this chosen tract, we determined that because of commuting to a tract with much higher emissions/concentrations of 1,3-butadiene, than in their rural home tract, modeled personal concentrations of some of these individuals were 10 times more than the corresponding 1,3-butadiene concentration levels in their home tract.

A direct comparison between HAPEM5 and personal monitoring data is not readily available. While the current version of HAPEM has not undergone a direct validation study, other probabilistic models similar to HAPEM have been compared to measurement data. MacIntosh et al. (1995) performed a preliminary validation exercise for the Benzene Exposure and Absorbed Dose Simulation (BEADS) model and found that the model produced reasonable estimates of the distribution of benzene personal air concentrations for large populations. Further, many of the key component of HAPEM, such as the activity data ME data, commuting data have undergone their own peer review (US EPA, 2001) thus increasing our confidence in the models predictions.

Like any modeling analysis, our analysis also has a number of limitations or is based on a number of assumptions. For example, the HAPEM model, especially when applied on a very broad geographical scale, relies on a number of simplifying assumptions and approximations in estimating inhalation exposures. Moreover, HAPEM calculates long-term average exposure concentrations to address exposures to pollutants with carcinogenic and other long-term effects. HAPEM only estimates exposures experienced through inhalation. For certain HAPs, inhalation might not be the major route of exposure, and therefore, HAPEM may underestimate exposures in these instances where multimedia contributions are important. In general, the accuracy of the results is limited by: the emissions inventory and dispersion modeling results; reliability of ME factors for various pollutants; activity-pattern information (e.g., patterns of time spent in various MEs for the populations in the geographic areas modeled); the ability of the chosen population cohorts to adequately represent the true demographics of every census tract; and the model algorithms formulations. More specifically, limitations and uncertainties of this approach include:

(1) Uncertainty with the emissions inventory and the ASPEN dispersion modeling, which was mentioned before.
(2) The exposure estimates do not include exposures related to indoor emission sources of HAPs (e.g., off-gassing

from building or consumer products, smoking, internal combustion sources, etc.), which will lead us to underestimate total exposures. Recent personal monitoring studies confirm the importance of the contributions from several VOCs and carbonyl sources in indoor residential MEs (Liu et al., 2006; Sexton et al., 2007). However, indoor sources could be addressed in the future as additional data are obtained and new analyses are conducted.

(3) The HAPEM model only estimates inhalation exposures, as it utilizes ambient air concentration data as inputs. Thus, it does not address exposures through other routes or pathways (e.g., ingestion). This is especially important for toxic pollutants that are persistent and bioaccumulate, such as mercury, dioxins, and PCBs. Emissions of these pollutants disperse through the atmosphere and eventually deposit to land or water bodies. Once deposited, they can bioaccumulate up the food chain. Multimedia exposure models are needed to address such multipathway exposures.

(4) The exposure estimates represent midrange estimates of population exposures. Due to a number of factors, such as variability in local ambient levels and activity patterns, some of the individuals may have substantially higher or lower exposures. This national scale assessment is not designed to quantify these extreme values of individual exposures.

(5) Exposure models must predict the relationship between the ambient air quality (outside) and that in an ME (inside or outside). When applied on a local scale, exposure models can employ detailed mass balance equations to predict this relationship. However, on a national scale, the development of such a detailed relationship is not feasible. Thus, the HAPEM model, applied on a national scale, relies on generalized ME factors, which do not account for variability (e.g., penetration affected by air exchange rate, which is a function of ambient temperature, heating/cooling system, open windows). As part of this assessment, Environmental protection Agency (EPA) conducted a detailed study of exposure literature to develop ME factors for each air toxic in the study, which has undergone a separate technical peer review (USEPA-SAB2002).

(6) The assessment assumed that the 10 demographic groups selected can represent the activity patterns of the general population in all areas of the country. The groups are selected to represent variability in population activity patterns while at the same time maintaining the ability to present the exposure assessment results in a manner that will allow for an adequate lifetime exposure aggregation.

(7) When selecting multiple 24-h activity patterns to construct an annual sequence, daily patterns are combined that pertain to different individuals, so that day-to-day correlations in activities are not preserved. For example, for day 1, the pattern may specify a house with an attached garage, and for day 2, a house without an attached garage. In this situation, the HAPEM model would underestimate the annual average exposure for a person residing in a house with an attached garage, and overestimate the exposure of the person in the house without an attached garage. As a result, the aggregated activity pattern is more representative of a population average pattern for the demographic group, than any individual pattern.

On-going or planned research activities by EPA are intended to address a number of these modeling limitations or challenges. For example, the EPA is planning on improving the emissions inventory that is used in NATA by leading a reengineering effort, in collaboration with State/Local/Tribal (S/L/T) agencies, to identify and implement process and technology improvements to: (1) create a more efficient way to develop the NEI and (2) to develop an NEI that is more accurate, timely, and transparent. Further, EPA is planning to begin integrating its air quality models by combining its Community Multi-scale Air Quality (CMAQ) model with a Gaussian model such as AERMOD to better predict both photochemical and long-range transport issues (Touma et al., 2006). Finally, the EPA is planning on making several improvements to its exposure modeling platforms. HAPEM will be upgraded to include the effects of living and working near-roadway environment. Other more refined exposure models such as APEX and SHEDS will continue to be used in special applications. Additional information on these improvements can be found on the agencies website. Based upon the results of this study, we also feel that further refinements in the information and methods used to predict worker- and school-commuting patterns would be highly beneficial.

In conclusion, we have determined that exposures to either near-roadway emissions or as a result of commuting to locations with greater pollution can significantly increase individuals exposures to outdoor HAPs, anywhere from 20% to even a factor of two or greater than the outdoor concentrations modeled at their home census tracts. These findings confirm some of the recent results on the important role of commuting and human activities in influencing population exposures to benzene and other motor-vehicle-related pollutants (Isakov et al., 2006; Marshall et al., 2006). Our analysis extends these results to many other gaseous and particulate HAPs and examines this issue further through a nationwide analyses by each of the five different emissions HAPs source category. We conclude that the increase in personal exposures from the corresponding ambient levels tends to occur near locations where there are either major emission sources of HAPs or when individuals are exposed to on- or nonroad sources of HAPs during their daily activities.

The implications of these findings to either the chronic or acute air pollution health effects studies of PM and HAPs can be quite important. Most of the air pollution epidemiology studies assign a fixed outdoor concentration value (either measured or modeled at the county or zip code of residence or at an individuals home address) as the exposure value (Burnett et al., 1998, 2000; Pope et al., 2002; McConnell et al., 2006; Domenici et al., 2006). This assignment introduces varying degrees of exposure prediction error depending on the nature of the epidemiologic model tested and the statistical methodology employed. We have shown that the variation in the exposure to concentration ratios can be highly pollutant, site and activity dependent. Thus, the complexity in the spatial variation of exposures among the different population cohorts, especially in the context of cross-sectional or intra-urban analysis of air pollution health effects, could be quite challenging. Recent advances in the development of more sophisticated exposure modeling tools and better information on time-activity, commuting and exposure factors data, should offer us a unique opportunity to improve the assignment of exposures during the course of future air pollution epidemiology studies.

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