Published in final edited form as:
Atmos Environ. 2011 October 1; 45(32): 5684–5691. doi:10.1016/j.atmosenv.2011.07.034.

Geographic differences in inter-individual variability of human exposure to fine particulate matter

Ye Cao and
Department of Civil, Construction and Environmental Engineering, North Carolina State University, Campus Box 7908, Raleigh, NC 27695-7908, (O) (919) 515-4232, (F) (919) 515-7908, ginnycao@gmail.com

H. Christopher Frey, Ph.D. [Professor]
Department of Civil, Construction and Environmental Engineering, North Carolina State University, Campus Box 7908, Raleigh, NC 27695-7908, (O) (919) 515-1155, (F) (919) 515-7908, frey@ncsu.edu

Abstract

Human exposure to fine particulate matter ($PM_{2.5}$) is associated with short and long term adverse health effects. The amount of ambient $PM_{2.5}$ that infiltrates indoor locations such as residences depends on air exchange rate (ACH), penetration factor, and deposition rate. ACH varies by climate zone and thus by geographic location. Geographic variability in the ratio of exposure to ambient concentration is estimated based on comparison of three modeling domains in different climate zones: (1) New York City; (2) Harris County in Texas, and (3) a six-county domain along the I-40 corridor in North Carolina. Inter-individual variability in exposure to $PM_{2.5}$ was estimated using the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model. ACH is distinguishably the most sensitive input for both ambient and nonambient exposure to $PM_{2.5}$. High ACH leads to high ambient exposure indoors but lower non-ambient exposure, and vice versa. For summer, the average ratio of exposure to ambient concentration varies by 13 percent among the selected domains, mainly because of differences in housing stock, climate zone, and seasonal ACH. High daily average exposures for some individuals are mainly caused by non-ambient exposure to smoking or cooking. The implications of these results for interpretation of epidemiological studies are discussed.

Keywords

exposure; particulate matter; variability; air exchange rate; penetration factor; deposition rate

1. Introduction

Fine particulate matter ($PM_{2.5}$) includes particles that are 2.5 microns or less in aerodynamic diameter. Exposure to $PM_{2.5}$ is associated with adverse health outcomes (EPA, 2009). Hence, there is a need to quantify human exposure to $PM_{2.5}$ to support assessment of its health effects. Individual exposures to $PM_{2.5}$ occur both outdoors and indoors, and indoor

$PM_{2.5}$ concentrations are affected by penetration of ambient $PM_{2.5}$ and exposures from sources such as cooking, cleaning and smoking (Lachenmyer and Hidy, 2000).

In recent epidemiology studies, associations between exposure to $PM_{2.5}$ and health effects are quantified as response-concentration functions based on multicity studies; however, exposure is not measured or estimated (EPA, 2009). These studies assumed that ambient concentration is a surrogate for exposure, but do not address whether the ratio of exposure to concentration is similar for different locations.

$PM_{2.5}$ exposure studies typically employ either direct measurement methods or estimate exposure using models. For example, Williams *et al.* (2003) performed a 1-year investigation in North Carolina of $PM_{2.5}$ and related co-pollutants to characterize the relationship between measured personal exposure versus ambient and residential $PM_{2.5}$ concentration. Mean daily personal $PM_{2.5}$ exposures were only moderately correlated to ambient $PM_{2.5}$ concentrations. Lachenmyer and Hidy (2000) conducted outdoor, indoor and personal exposure measurements for a sample population in Alabama and observed a weakly linear relationship between personal exposure and ambient $PM_{2.5}$ concentration.

Population-based exposure monitoring is an economical tool for quantifying personal exposure but requires considerable resources In contrast, scenario-based exposure models estimate personal exposure for simulated members of a defined population. based on the time spent in specific microenvironments, including home, school, store, restaurant and vehicles (Burke *et al.,* 2001). Total individual exposure is calculated from the sum of the microenvironmental exposures over the course of an averaging time of interest, such as a typical weekday. As an example, the Stochastic Human Exposure and Dose Simulation for Particulate Matter (SHEDS-PM) model, developed by the US Environmental Protection Agency (EPA), uses a probabilistic approach that incorporates to estimate distributions of outdoor and indoor $PM_{2.5}$ exposure for a population of simulated individuals based on ambient $PM_{2.5}$ concentrations and sources of indoor $PM_{2.5}$ emissions (Burke, 2005).

SHEDS-PM inputs include demographic data, ambient $PM_{2.5}$ concentration, and human activity data. Demographic data are from the 2000 U.S. Census. The daily average ambient $PM_{2.5}$ concentration for each census tract for the geographic area of interest can be based on ambient monitoring or air quality modeling data such as from the Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006). The amount of time each person spends during a typical day in each microenvironment is quantified based on the Consolidated Human Activity Database (CHAD), which is comprised of U.S. human activity pattern diary data compiled based on multiple activity studies (Johnson 1984, 1989; Settergren *et al.,* 1984; Wiley 1991; Klepeis *et al.,* 1996; McCurdy *et al.,* 2000).

The key factors affecting the fraction of ambient particles that penetrate indoors and remain suspended are: (1) air exchange rate (ACH); (2) penetration factor (P); and (3) deposition rate (k) (Wilson *et al.,* 2000). ACH is estimated based on measurements with a tracer gas, such as perfluorocarbon tracer (PFT) or sulfur hexafluoride (SF_6). P and k are difficult to measure directly, but are typically estimated by fitting a mass balance model to data for paired indoor and outdoor concentration and ACH. Few observational data are available on seasonal and geographic variability P and k.

The estimated exposure (E) can be conceptualized as a linked source-to-exposure model by the coupling of daily average ambient concentration (C) from an output of an air quality model and an exposure model that estimates the ratio of exposure to ambient concentration (E/C) (Özkaynak *et al.*, 2009). The ratio E/C is approximately independent of C for ambient sources of exposure, and varies geographically depending on demographics and housing stock, and infiltration parameters ACH, P and k (Burke, 2005). Thus, E/C may vary seasonally among geographic areas.

The objectives of this paper are to: (1) review and recommend values of ACH, P, and k for selected geographic areas; (2) conduct sensitivity analysis for ACH, P, and k to evaluate their importance; (3) evaluate geographic differences in inter-individual variability in exposure; and (4) evaluate geographic differences in the ratio of exposure to concentration.

2. Methodology

The methodology includes: (1) review of the algorithm and values ACH, P, and k for estimating residential $PM_{2.5}$ microenvironmental concentration; (2) sensitivity analysis of ACH, P, and k to assess their importance with respect to estimated exposure; (3) characterization of geographical variability associated with total daily average $PM_{2.5}$ exposure; and (4) characterization of the ratio of exposure to ambient concentration for ambient exposure, non-ambient exposure, and total exposure in each geographic area.

2.1 Residential $PM_{2.5}$ concentration

SHEDS-PM includes a single-compartment, steady-state mass balance equation to estimate the indoor $PM_{2.5}$ concentration in the residential microenvironment (Burke *et al.* 2001). Indoor residential $PM_{2.5}$ includes outdoor PM that enters indoors and PM generated by indoor emission sources such as cigarette smoking, cooking, and cleaning:

$$C_{Home}=\frac{P\cdot ACH}{ACH+k}C_{ambient}+\frac{\Sigma E_i}{(ACH+k)VT} \quad (1)$$

Where,

ACH = air exchange rate (h^{-1});

C_{Home} = $PM_{2.5}$ concentration in the home ($\mu g/m^3$);

$C_{ambient}$ = ambient outdoor $PM_{2.5}$ concentration ($\mu g/m^3$);

E_i = emissions from indoor sources i;

k = deposition rate (h^{-1});

N_{cig} = number of cigarettes smoked during model time step (cig);

P = penetration factor (unitless);

T = model time step (min);

V = volume of microenvironment (m^3).

ACH, P, and k can be specified as probability distributions. ACH is the volume flow of air within the indoor microenvironment divided by the interior volume. ACH is affected by air leakage through cracks and crevices in the building envelope, natural ventilation through open windows and doors, and mechanical ventilation by fans (Liu and Nazaroff, 2001).

SHEDS-PM categorizes ACH into four seasons: winter, spring, summer, and fall. The default data for ACH for these seasons was originally derived from a PFT database developed by Brookhaven National Laboratory (BNL). Murray and Burmaster (1995) analyzed the database and categorized ACH by climate region and season. However, regional variations of ACH represented in Murray and Burmaster (1995) are not included in SHEDS by default.

NIH-PA Author Manuscript

NIH-PA Author Manuscript

P is the ratio of particles that penetrate indoors from outdoors. k refers to settling of airborne particles due to gravity and diffusion. The deposition rate depends on particle size source strength and ventilation conditions (He *et al.,* 2005; Thatcher *et al.,* 2002; Lai and Nazaroff, 2000). The default values of P and k in SHEDS were obtained from the Particle Total Exposure Assessment Methodology (PTEAM) study conducted for Riverside, California, in fall of 1990 (Özkaynak *et al.* 1997).

2.2 Review of penetration, depostion, and air exchange rates

The review of P, k, and ACH is based on: (a) detailed review of SHEDS-PM, its user guide, and the literature cited as the basis for default input assumptions; (b) published peer reviewed papers regarding similar models; and (c) published peer reviewed papers regarding data for ACH, P and k. Data are reviewed with respect to selected geographic areas and for four seasons.

2.3 Sensitivity analysis

Sensitivity analysis of an exposure model helps to identify the most significant factors that aid in risk management or that enable prioritization of additional research to reduce uncertainty in the estimates (Frey and Patil, 2002). Sensitivity analysis was conducted to assess the variability in daily average $PM_{2.5}$ exposure as a function of variation in P, k, and ACH.

During the sensitivity analysis, all inputs were held at their default values except for one, which was varied probabilistically. Results are shown as a Cumulative Distribution Function (CDF) of inter-individual variability in daily average exposure for simulated individuals. Based on the percent difference in the mean and standard deviation of exposure associated with comparison of alternative distributions for each selected input, the key inputs were identified and prioritized.

2.4 Geographic and inter-individual variability

To assess the geographic variability in estimated exposure, three locations were selected that represent different climate zones: (1) New York City (NYC); (2) Wake, Durham, Orange, Alamance, Guilford, and Forsyth Counties in North Carolina, which includes the cities of Raleigh, Durham, Burlington, Greensboro, High Point, and Winston-Salem; and (3) Harris County in Texas, which includes Houston. Since the average ambient $PM_{2.5}$ concentration tends to be highest in the summer, air quality data for July 2002 were selected. The six counties selected for the NC case study represent urban areas along the I-40 highway corridor.

SHEDS-PM output includes a database for each individual for each simulated day, with estimates of daily average microenvironmental exposure concentrations for ambient, nonambient, and total exposure. The ratio of exposure to ambient concentration for ambient exposure, non-ambient exposure, and total exposure in each geographic area are based on the pairwise estimated exposure and assigned ambient concentration for each simulated individual.

3. Results

Recommended values of ACH, P, and k are given. Data regarding ACH are reviewed mainly based on Murray and Burmaster (1995) and the Relationship of Indoor, Outdoor and Personal Air (RIOPA) study from 1999 to 2001. Data regarding P and k are reviewed mainly based on the RIOPA and PTEAM Studies (Weisel *et al.* 2005; Özkaynak *et al.*

1997). Each input is evaluated using sensitivity analysis. Geographic and inter-individual variability in daily average exposure to $PM_{2.5}$ is evaluated for summer.

3.1 Air exchange rate

Murray and Burmaster (1995) summarized ACH data compiled by BNL for 2,844 households. This is the most recently reported comprehensive analysis of such data. They stratified the data into four regional climate zones based on heating degree days. For each region and season, lognormal distributions were fit to the data to represent inter-household variability. New York State includes two regions, and NYC is in region 2. Koontz and Rector (1995) analyzed 2,976 measurement results from the BNL database stratified by state rather than climate zone.

Summer estimates for Region 2 are based mostly on data from Washington State, and thus are not representative of NYC (Murray and Burmaster, 1995). Koontz and Rector (1995) report geometric mean (μ_g) for New York State but not geometric standard deviation (σ_g). However, Murray and Burmaster (1995) report σ_g of 2.09 for Region 2, which includes New York State just north of NYC. Thus, ACH for summer in NYC is assumed to have μ_g of 0.64 with σ_g of 2.09.

For summer and Texas, μ_g and σ_g from the RIOPA and Koontz and Rector (1995) data are much lower than from Murray and Burmaster (1995). Because air conditioning usage is higher in Texas than in the northern regions (EIA, 2000), ACH is expected to be lower in the summer than for other seasons. The seasonal trend in μ_g from the RIOPA data is internally constant and thus these data (μ_g = 0.37, σ_g=1.90) are used for summer.

Wallace *et al.* (2006) measured ACH in 37 residences in the Research Triangle Park (RTP) area in North Carolina for 7 consecutive days in each of four seasons. The highest mean value of ACH was observed in the winter and the lowest in the summer, a reversal of the typical pattern in other studies (Abt *et al.,* 2000; Long *et al.,* 2001). North Carolina is in Region 3 in Murray and Burmaster (1995). For summer, both μ_g and σ_g from Murray and Burmaster (1995) are similar to these reported by Wallace *et al.* (2006). The data from Wallace *et al.* (2006) (μ_g = 0.54, σ_g=1.70) are used for summer.

3.2 Penetration factor and deposition rate

Ideally, P and k should be measured and estimated for houses with no indoor emission sources. However, many of the available data appear to be for houses with indoor sources, except as noted. The presence of indoor sources will produce bias in P, including values greater than 1, and bias in k. In some cases, estimates of k are negative.

In the RIOPA study, there are a limited number of households (n=21) for which there is explicit reporting of no indoor emission sources (Weisel *et al.,* 2005). For these households, P and k are 0.78, and 0.40 h^{-1}, respectively. For 165 households, the indoor concentration was less than the outdoor concentrations, denoted as I/O<1. I/O<1 is consistent with houses that do not have indoor sources, but it is not adequate assurance that no indoor sources were present. For these data, the average values of P and k are 0.73 and 0.20 h^{-1}, respectively. Data for other RIOPA locations may include indoor sources and, thus, might be biased. The PTEAM study was conducted in Riverside, California in fall 1990. Indoor concentrations were measured in the center of the residence and outdoor concentrations were measured nearby. Measurements were conducted daily for two consecutive 12-h periods. During each 12-h period, PFT measurements were made from which ACH was estimated.

Based on the physical constraint that $P \le 1$, an upper bound of P = 1 is assumed. A lower bound of 0.7 is assumed consistent with the mean value observed for the RIOPA study for I/

O<1, Elizabeth, NJ, and the lower bound of the reported 95 percent Confidence Intervals (CI)s for Los Angeles and the overall data. A nominal "best estimate" of 0.78 is assumed based on the mean of the data for which no indoor sources were reported. A triangular distribution is used for P to represent these judgments. Triangular distributions are sometimes used to represent judgment in the absence of a probability sample of data (Cullen and Frey, 1999).

A lower bound of k = 0.2 h^{-1} is assumed consistent with the mean value observed for the RIOPA data in which I/O ≤ 1, the lower bound of reported CIs for overall cases, and the lower bound of the CI for Los Angeles from the PTEAM study. A nominal "best estimate" of 0.40 h^{-1} is assumed based on the mean of data for which no indoor sources were reported, and the mean of data from the PTEAM study. The upper bound of k = 0.6 h^{-1} is assumed based on the upper bound of the CI from the PTEAM study. A normal distribution is assumed for k with a mean of 0.40 h^{-1} and standard deviation of 0.1, giving a 95 percent probability range of approximately 0.2 to 0.6 h^{-1}.

3.3 Sensitivity analysis

The specified distributions for these inputs in sensitivity analysis are summarized in Table 1. In addition to a triangular distribution of P, two alternative frequency distributions are also considered: (1) normal distribution with a mean of 0.85 and a standard deviation of 0.075; (2) normal distribution with a mean of 0.90 and a standard deviation of 0.05. An alternative normal distribution is also considered for k with a mean of 0.39 h^{-1} and a standard deviation of 0.085, which are consistent with the data observed for the PTEAM study.

To compare the sensitivity of estimated exposure to each of several residential microenvironmental inputs, a case study was developed based on all census tracts in Harris County, Texas. For each census tract, 1% of the population was simulated. Distributions for P and k are assumed to be the same for the three geographic areas, while ACH differs. Sensitivity analysis for ACH is conducted for summer using air quality data for July 2002. These data are estimated based on the predictions of average concentrations for 12 by 12 km grid cells obtained from CMAQ and combined, using Bayesian statistical methods, with monitoring data (McMillan *et al.,* 2010). Model validation was analyzed by the comparison between predictions from kriging model based solely on monitoring data and Bayesian model. The Bayesian model and kriging approaches provided similar mean squared prediction error across all Bayesian model runs. The Bayesian model reduced bias by 10 to 15 percent compared to kriging (McMillan *et al.,* 2010). However, $PM_{2.5}$ concentration tends to have relatively less spatial variability at a given time than other pollutants (EPA, 2009). Thus, the use of monitoring data in a Bayesian framework to "update" the CMAQ predictions will improve the accuracy of the representation of data for a region at a given time. Furthermore, updating based on monitoring data helps ensure better representation of temporal trends in concentration.

All exposure model runs were conducted using longitudinal simulations and the same random seed. Five simulations were conducted for various combinations of probabilistic assumptions for P, k, and ACH: (1) P_1, k_1, ACH_1; (2) P_2, k_1, ACH_1; (3) P_3, k_1, ACH_1; (4) P_1, k_2, ACH_1; and (5) P_1, k_1, ACH_2. Each model run was conducted on a Windows XP Pentium 4 computer and had an approximate runtime of 800 minutes.

The three alternative distributions for P differ in terms of central tendency and range. However, the corresponding three simulated distributions of inter-individual variability in exposure are very similar. The mean exposure differs by less than 5 percent and the standard deviations differ by less than 2 percent. Therefore, the results are not sensitive to the choice of distribution for P. Since P_3 is the only one of the three that observes the physical limit of $P \leq 1$, P_3 is chosen as the basis for further simulations.

The two alternative distributions for k differ in terms of central tendency and. However, the corresponding simulated distributions of inter-individual variability in exposure are very similar. Both the mean exposure and the standard deviation of exposure differ less than 1 percent. Therefore, the results are not sensitive to the choice of distribution, and k_1 is chosen as the basis for further simulations.

The two alternative distributions of ACH for Texas differ in terms of central tendency and range as shown in Fig. 1(a). The μ_g and σ_g of ACH_1 are 65 and 24 percent higher than those of ACH_2, respectively. The corresponding simulated distributions of inter-individual variability in mean exposure and standard deviation based on ACH_1 is 67 and 40 percent lower than that based on ACH_2 as shown in Fig. 1(b), respectively. Therefore, the results are sensitive to the choices of distributions of ACH. ACH_2 is chosen because it is consistent with the expected seasonal trends as explained in Section 3.1.

3.4 Geographic variability of exposure to $PM_{2.5}$

A random sample of 50,000 individuals in all age groups from all census tracts in each of NYC, Harris County in Texas, and the six counties in North Carolina was simulated to characterize the geographic variability of estimated exposure to $PM_{2.5}$. $PM_{2.5}$ air quality data are the same as used in sensitivity analysis. The average daily ambient $PM_{2.5}$ concentration was 20.9, 15.7, and 20.6 $\mu g/m^3$ in the NY, TX, and NC domains, respectively. Longitudinal simulation was used in all model runs.

Inputs that vary among geographic areas, including ACH, smoking prevalence, demographics, and distribution of housing types, are given in Table 2. A detailed review of smoking prevalence data and its adequacy is given by Cao and Frey (2011). Because of lack of county level smoking prevalence data, state level data were used. The central tendency of summer ACH in Texas is 31 and 42 percent lower than for the NC and NYC domains, respectively. Lower ACH leads to more retention of indoor emissions, and higher non-ambient exposure. Conversely, lower ACH leads to lower penetration of ambient $PM_{2.5}$ indoors, and lower ambient exposure.

Based on the distribution of smoking by gender and age, and the population distribution by age and gender, the weighted overall prevalence of smokers are 23.4, 20.0, and 19.1 percent for NC, TX, and NYC, respectively. The smoking prevalence varies substantially among different age and gender cohorts. For example, the proportion of smokers older than 45 years old is 28 percent higher than those in other age groups in Harris County, TX. Furthermore, the proportion of time spent indoors also varies by cohort. Based on CHAD diary data, the distribution of daily average time spent outdoors, indoors, and in travel by gender and age are given in Table 3. The time spent indoors for people older than 64 years is 16 percent higher than that of people from 14 to 15 years old. Differences in human activity patterns lead to difference in exposures.

The average interior volume of residential housing is approximately similar for the NC and TX domains, but approximately 30 percent lower in NYC. The smaller average housing interior volume is because the proportion of multiple family (apartment) housing in NYC is more than 50 percentage points larger than that in North Carolina. The NC and TX domains have approximately two-thirds single family houses, which have larger interior volume than apartments. Large indoor volume enhances the dilution of indoor emissions. Smaller indoor volume leads to higher indoor exposure to non-ambient $PM_{2.5}$.

When the activity patterns in Table 3 are weighted based upon the geographic location-specified population distribution, as given in Table 2, differences in activity patterns between cohorts can be pronounced in some cases. For example, even though the same activity diary data are used, because there is a larger proportion of 18 to 24 year old males in NC, the overall proportion of time spent outdoors by 18 to 24 year old males in NC is 4 and 13 percent more than those for TX and NYC, respectively. The aggregated proportion of time spent outdoors for people older than 45 years old in NYC is estimated to be 6 percent more than those of TX and NC domains, respectively. There is also a larger proportion of smokers older than 45 years old in NYC.

The estimated daily average ambient exposure in NYC is 12.7 $\mu g/m^3$, with a 95 percent frequency range, as shown in Fig. 2(a), of 3.4 to 24.6 $\mu g/m^3$. For Harris County, the mean is 8.4 $\mu g/m^3$, with a range of 3.4 to 14.1 $\mu g/m^3$. For the six county area of North Carolina, the mean is 12.2 $\mu g/m^3$, with a range of 4.4 to 21.0 $\mu g/m^3$.

The higher estimated ambient exposure in NYC is attributed in part to higher ambient $PM_{2.5}$ concentration, which averages 25 and 1.4 percent higher than that of Texas, and North Carolina, respectively. The higher summer ACH in NYC leads to more indoor penetration of ambient $PM_{2.5}$. Ambient exposure for NC is 31 percent higher than that for TX. This is attributed in part to higher ACH and ambient $PM_{2.5}$ concentration in NC than for Harris County.

The average estimated non-ambient exposure in Harris County is 14 and 11 percent higher than that of the NC and NY domains. The population weighted smoking prevalence in Texas is lower than that of NC. The weighted average indoor residential volume for Texas is higher than that of NC and NYC. These two factors lead to lower non-ambient exposure to $PM_{2.5}$. However, the low summer ACH in Texas leads to overall higher non-ambient exposure. This implies that non-ambient exposure is more sensitive to ACH than to other factors, at least in this comparison.

NYC has a lower weighted average smoking prevalence and higher summer ACH than NC, which would favor lower non-ambient exposure. However, the smaller average indoor volume in NYC leads to overall higher non-ambient exposure to $PM_{2.5}$. This implies that estimated nonambient exposure is sensitive to the distribution of housing stock.

The mean values of total daily average exposure to $PM_{2.5}$ in the NYC, NC, and TX domains are 28.9, 27.8, and 26.6 $\mu g/m^3$, respectively. The comparative order of these averages is consistent with the order of average ambient exposure. However, the 90th percentile of total exposure is higher in Harris County by 3 and 7 percent compared to NYC and NC, respectively. The latter trend is consistent with the trend in non-ambient exposure. Thus, geographic variability of total daily average exposure is influenced by the variability in ambient exposure. However, the geographic variation in high-end exposure is influenced by variability in nonambient exposure. This is because the high-end of non-ambient exposures can be a factor of 2 to 10 higher than the ambient exposures. The high-end exposures are mainly caused by indoor emissions, such as, smoking and cooking. The estimated average non-ambient exposures for smokers are approximately fivefold higher than those for non-smokers in each on the three geographic areas.

3.5 Inter-individual variability in exposure to $PM_{2.5}$

Fig. 3 illustrates variability in the ratio of estimated exposure to ambient concentration for ambient exposure (E_a/C), non-ambient exposure (E_{na}/C), and total exposure (E_t/C). The estimated mean values of Ea/C given in Fig. 3(a) are 0.60, 0.58, and 0.52, with standard deviations of 0.16, 0.15, and 0.14, for the NY, NC and TX domains, respectively. The geographic differences in the mean Ea/C ratio are mainly associated with differences in average ACH.

NYC has the highest E_a/C ratio. Higher ACH in NYC leads to higher penetration of ambient $PM_{2.5}$ indoors, which leads to higher exposure to ambient $PM_{2.5}$. The average E_a/C in the three geographic areas increase as the mean ACH increases. Each of the average, 50th percentile, and 90th percentile of E_a/C varied approximately by 3 percent between the NYC and NC domains, 9 percent between the NC and TX domains, and 12 percent between the NYC and TX domains. The 50th percentile of ACH in NYC is 16 and 42 percent higher than those of the NC and TX domains, the 90th percentile of ACH in NYC is 2 and 13 percent higher than those the TX and NC domains. The relative magnitude of E_a/C appears to be insensitive to population demographics when comparing the three geographic areas. For example, the comparative increase in mean E_a/C for NYC, NC, and TX does not correspond to a demographic trend such as the proportion of the population aged 18 to 65.

Based on Fig. 3(b), the estimated average ratios of E_{na}/C are 1.2, 1.0, and 0.9, with standard deviations of 2.1, 1.8, and 1.4, for the TX, NY and NC domains, respectively. Geographic variability in E_{na}/C is associated with ambient air quality, ACH, smoking prevalence, and indoor volume. The TX domain has the lowest ACH and the lowest average ambient air quality. Low ACH leads to more retention of indoor emissions. Thus, TX has the highest average E_{na}/C ratio. The overall smoking prevalence in TX is lower than that of NC. The proportion of single family homes in TX is 67 percentage points more than that of NYC; therefore, the average indoor volume of TX is greater than that of NYC. Larger indoor volume helps the dilution of indoor emission sources, such as smoking and cooking.

Based on Fig. 3(c), the estimated average ratios of E_t/C are 1.8, 1.6, and 1.4, with standard deviations of 2.1, 1.8, and 1.5, for the TX, NY and NC domains, respectively. The relative magnitudes of the average and 90th percentile E_t/C ratios among geographic areas are consistent with the order of the average E_{na}/C ratio. However, at the 50th percentile, E_t/C is consistent with the order of the E_a/C ratio. The 50th percentile of the E_t/C ratio for NYC is 17 and 35 percent higher than for the TX and NC domains, respectively. This indicates that the average personal exposures to $PM_{2.5}$ are dominated by ambient exposure; however, the high-end personal exposures are more influenced by non-ambient exposure.

4. Conclusions

ACH is distinguishably the most sensitive input for both ambient and non-ambient exposure to $PM_{2.5}$. The distributions of E_a/C ratios in each geographic area indicate that simulated individuals are typically exposed to more than half of the modeled ambient concentrations. The estimated average E_a/C ratio relatively varies 13 percent among geographic areas based on differences in housing stock, climate zone, and seasonal ACH. However, the range of inter-individual variability in exposures tends to be similar. High ACH, whether on average for a geographic area or for a specific individual regardless of geographic area, leads to high ambient exposure indoors but lower non-ambient exposure, and vice versa. Thus, geographic areas or seasons with higher average values of ACH are likely to have relatively higher average ratios of ambient exposure to ambient concentration. One implication is that assessments of human exposure and risk to $PM_{2.5}$ should identify regions and seasons with high ACH as being potentially highly exposed and thus prioritized for further investigation.

Approximately 60 percent of the estimated total daily average exposures to $PM_{2.5}$ are caused by non-ambient exposures for each of the three geographic areas. The extremely high daily average exposures for some individuals are mainly caused by non-ambient exposure to smoking or cooking. The magnitude of non-ambient exposure for a given indoor source strength is inversely proportional to ACH and to interior housing volume. Indoor behavior and air quality at home is typically treated as a private matter. Thus, these results imply a need to develop communications to the public to increase awareness of the potentially very high levels of indoor particles that occur particularly as a result of smoking and some kinds of cooking, to encourage voluntary measures at prudent avoidance of such exposures.

Geographic differences in housing stock and climate, which lead to geographic differences in ACH, account for geographic variability in the estimated ambient and non-ambient exposure to $PM_{2.5}$. The microenvironmental simulation modeling approach can take into account region- and climate-specific values of these key inputs, and can be applied more broadly than demonstrated here to provide additional insights regarding geographic and seasonal differences in exposures.

There are a number of limitations of this work that motivate priorities for future refinement of the input data used for microenvironmental exposure modeling of $PM_{2.5}$. For example, there are relatively few data from which to estimate P and k. Data are needed for P and k that more clearly represent situations without indoor emissions. Because P and k are likely to be different among $PM_{2.5}$ species, further studies are required to obtain P and k for different $PM_{2.5}$ species. Furthermore, more recent data for ACH are needed to represent current housing stock in different regions. Although CHAD contains 22,968 diaries, there are not sufficient geographic or seasonal coverage of these diaries to specifically represent a given location or season. There is a need for more extensive diary data to represent multiple climate zones and land use patterns.

Most epidemiology studies use central site ambient monitoring data as a surrogate for population-level exposure. The estimates produced here imply that, on average, exposure concentrations are 50 to 80 percent of ambient concentrations, and that there is substantial inter-individual variability in exposures for a given ambient concentration. Because the average Ea/C ratio is significantly less than one, the effects estimates from epidemiology studies are biased low if exposure, rather than ambient air quality, is taken into account. Furthermore, there are differences in estimated E_a/C ratios that are attributed to geographic differences in ACH, housing distribution, and population distribution. Factors such as these could lead to inter-city variations in concentration-response functions inferred using epidemiological methods. Area-specific E_a/C ratios are recommended for possible use in epidemiology studies to better address geographic and inter-individual variability in exposure to $PM_{2.5}$.

Despite limitations of scenario-based exposure modeling associated with the current scarcity of data for factors such as ACH and diaries, the results obtained are plausible. The potential use of scenario-based exposure models to aid in explaining or quantifying inter-city variations in dose-response functions merits further attention.

References

Abt E, Suh HH, Catalano P, Koutrakis P. Relative contribution of outdoor and indoor particle sources to indoor concentrations. Environmental Science & Technology. 2000; 34:3579–3587.

Burke JM, Zufall MJ, Ozkaynak H. A population exposure model for particulate matter: case study results for $PM_{2.5}$ in Philadelphia, PA. Journal of Exposure Analysis and Environmental Epidemiology. 2001; 11:470–489. [PubMed: 11791164]

Burke, JM. SHEDS-PM stochastic human exposure and dose simulation for particulate matter user guide EPA Sheds-PM 2. 1. Washington, DC: United States Environmental Protection Agency; 2005. EPA/600/R-05/065

Byun D, Schere KL. Review of the governing equations, computational algorithms, and other components of the models-3 Community Multiscale Air Quality (CMAQ) modeling system. Applied Mechanics Reviews. 2006; 59:51–77.

Cao Y, Frey HC. Assessment of Inter-Individual and Geographic Variability in Human Exposure to Fine Particulate Matter in Environmental Tobacco Smoke. Risk Analysis. 2011; 31:578–591. [PubMed: 21039708]

Cullen, AC.; Frey, HC. The Use of Probabilistic Techniques in Exposure Assessment: A Handbook for Dealing with Variability and Uncertainty in Models and Inputs. New York: Plenum; 1999. p. 72

EIA. Trends in residential air-conditioning usage from 1978 to 1997. U.S. Energy Information Administration; 2000.

EPA. Intergrated Science Assessment for Particulate Matter. Washington, DC: United States Environmental Protection Agency; 2009. (Final Report). EPA/600/R-08/139F

Frey HC, Patil SR. Identification and review of sensitivity analysis methods. Risk Analysis. 2002; 22:553–578. [PubMed: 12088234]

He C, Morawska L, Gilbert D. Particle deposition rates in residential houses. Atmospheric Environment. 2005; 39:3891–3899.

Johnson, T. Study of Personal Exposure to Carbon Monoxide in Denver, Colorado. Research Triangle Park, NC: U.S. Environmental Protection Agency; 1984. EPA 1.89/2-600/S4-84-014, Prepared by Environmental Monitoring Systems Laboratory

Johnson, T. Human activity patterns in Cincinnati, Ohio, Final Report. Palo Alto, CA: Prepared for Electric Power Research Institute, Health Studies Program; 1989.

Klepeis, NE.; Tsang, AM.; Behar, JV. Analysis of the National Human Activity Pattern Survey (NHAPS) Respondents from a Standpoint of Exposure Assessment. Washington, DC: U.S. Environmental Protection Agency; 1996. EPA600/R-96-074

Koontz, MB.; Rector, HE. Estimated of distribution of residential air exchange rates. Washington, DC: United States Environmental Protection Agency; 1995. EPA 600/R-95/180

Lachenmyer C, Hidy GM. Urban measurements of outdoor-indoor $PM_{2.5}$ concentrations and personal exposure in the deep south. Part I. Pilot study of mass concentrations for nonsmoking subjects. Aerosol Science and Technology. 2000; 32:34–51.

Lai ACK, Nazaroff WW. Modeling indoor particle deposition from turbulent flow onto smooth surfaces. Journal of Aerosol Science. 2000; 31:463–476.

Liu DL, Nazaroff WW. Modeling pollutant penetration across building envelopes. Atmospheric Environment. 2001; 35:4451–4462.

Long CM, Suh HH, Catalano PJ, Koutrakis P. Using time- and size-resolved particulate data to quantify indoor penetration and deposition behavior. Environmental Science & Technology. 2001; 35:2089–2099. [PubMed: 11393992]

McCurdy T, Glen G, Smith L, Lakkadi Y. The National Exposure Research Laboratory's consolidated human activity database. Journal of Exposure Analysis and Environmental Epidemiology. 2000; 10:566–578. [PubMed: 11140440]

McMillan NJ, Holland DM, Morara M, Feng JY. Combining numerical model output and particulate data using Bayesian space-time modeling. Environmetrics. 2010; 21:48–65.

Murray DM, Burmaster DE. Residential air exchange-rates in the United States empirical and estimated parametric distributions by season and climatic region. Risk Analysis. 1995; 15:459–465.

NCHS. Health, United States, 2007 with chartbook on trends in the health of Americans. Hyattsville, MD: National Center for Health Statistics; 2007.

Özkaynak H, Frey HC, Burke J, Pinder RW. Analysis of coupled model uncertainties in source-to-dose modeling of human exposures to ambient air pollution: A $PM_{2.5}$ case study. Atmospheric Environment. 2009; 43:1641–1649. [PubMed: 20041038]

Özkaynak, H.; Xue, J.; Weker, R.; Butler, D.; Koutrakis, P.; Spengler, J. The Particle Team (PTEAM) Study: Analysis of the Data. Research Triangle Park, NC: United States Environmental Protection Agency; 1997. EPA/600/SR-95/098

SAMHSA. Results from the 2007 national survey on drug use and health: national findings. Rockville, MD: Substance Abuse and Mental Health Services Administration, Office of Applied Studies; 2008. NSDUH Series H-34 DHHS Publication No. SMA 08-4343

Settergren, SK.; Hartwell, TD.; Clayton, CA. Study of Carbon Monoxide Exposure of Residents of Washington, DC: Additional Analyses. Research Triangle Park, NC: Prepared for U.S. Environmental Protection Agency, Environmental Monitoring Systems Laboratory; 1984.

Thatcher TL, Lai ACK, Moreno-Jackson R, Sextro RG, Nazaroff WW. Effect of room furnishings and air speed on particle deposition rates indoors. Atmospheric Environment. 2002; 36:1811–1819.

Weisel, CP.; Zhang, J.; Turpin, BJ.; Morandi, MT.; Colome, S.; Stock, TH.; Spektor, DM.; Korn, L.; Winer, AM.; Kwon, J.; Meng, QY.; Zhang, L.; Harrington, R.; Liu, W.; Reff, A.; Lee, JH.; Alimokhtari, S.; Mohan, K.; Shendell, D.; Jones, J.; Farrar, L.; Maberti, S.; Fan, T. Relationships of indoor, outdoor, and personal air (RIOPA): Part I. Collection methods and descriptive analyses. Boston, MA: Health Effects Institute; 2005. (Report No. HEI Research Report 130)

Wallace, LA.; Williams, RW.; Suggs, J.; Jones, PA. Estimated Contributions of Outdoor Fine Particles to Indoor concentrations and Personal Exposures: Effects of Household Characteristics and Personal Activities. Research Triangle Park, NC: United States Environmental Protection Agency; 2006. EPA/600/R-06/023

Wiley, J. The study of children’s activity patterns, Final Report. Sacramento, CA: Prepared for California Air Resources Board, Research Division; 1991.

Williams R, Suggs J, Rea A, Leovic K, Vette A, Croghan C, Sheldon L, Rodes C, Thornburg J, Ejire A, Herbst M, Sanders W Jr. The Research Triangle Park particulate matter panel study: PM mass concentration relationships. Atmospheric Environment. 2003; 37:5349–5363.

Wilson WE, Mage DT, Grant LD. Estimating separately personal exposure to ambient and non-ambient particulate matter for rpidemiology and tisk Asssessment: ehy and how. Journal of the Air and Waste Management Association. 2000; 50:1167–1183. [PubMed: 10939210]

Research Highlights

- Most human exposure to ambient fine particulate matter ($PM_{2.5}$) occurs indoors
- Ambient exposure to concentration (E_a/C) for $PM_{2.5}$ averages 0.52 to 0.60
- Average E_a/C varies with geographic differences in residential air exchange rates
- The coefficient of variation of inter-individual variability in E_a/C is 0.25
- Variations in E_a/C are not accounted for in many health effects studies.

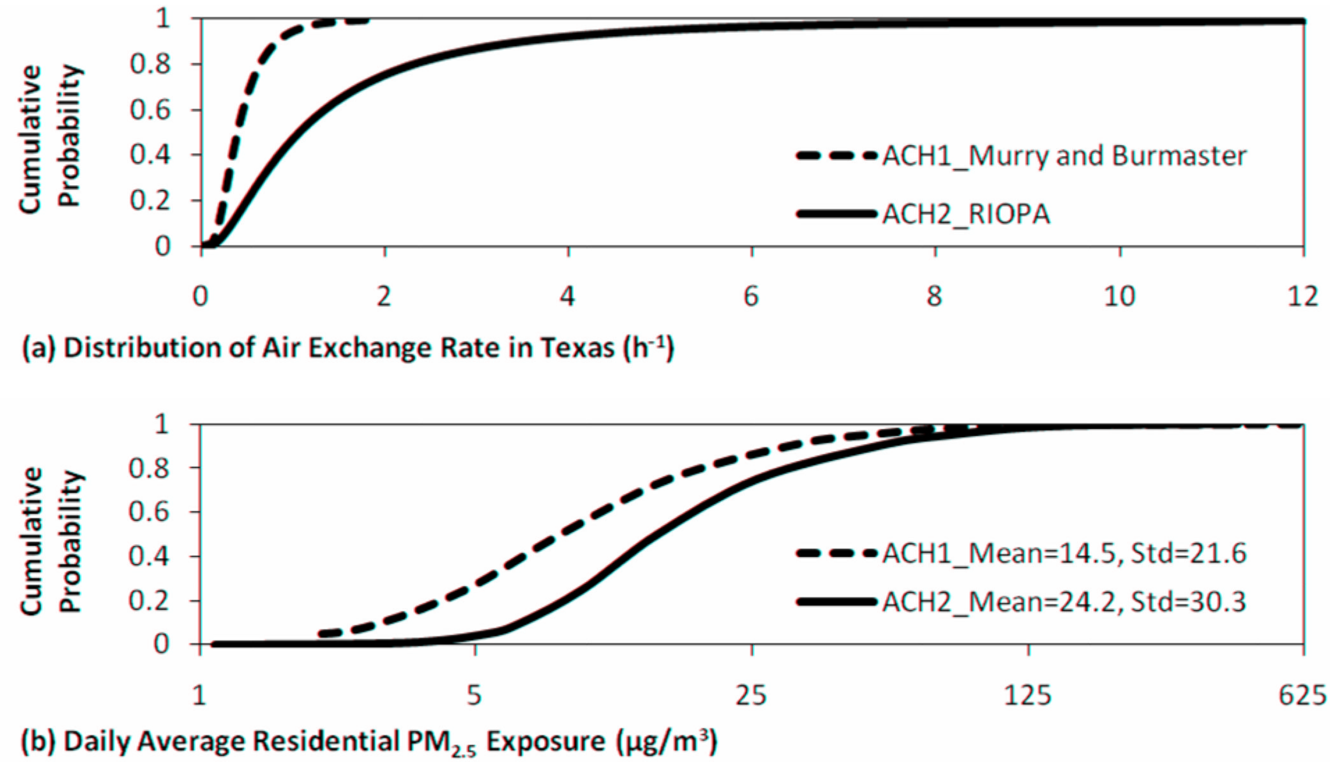

Fig. 1.

Variability in air exchange rate and daily residential $PM_{2.5}$ exposure for Harris County, TX for July 2002

NIH-PA Author Manuscript
NIH-PA Author Manuscript

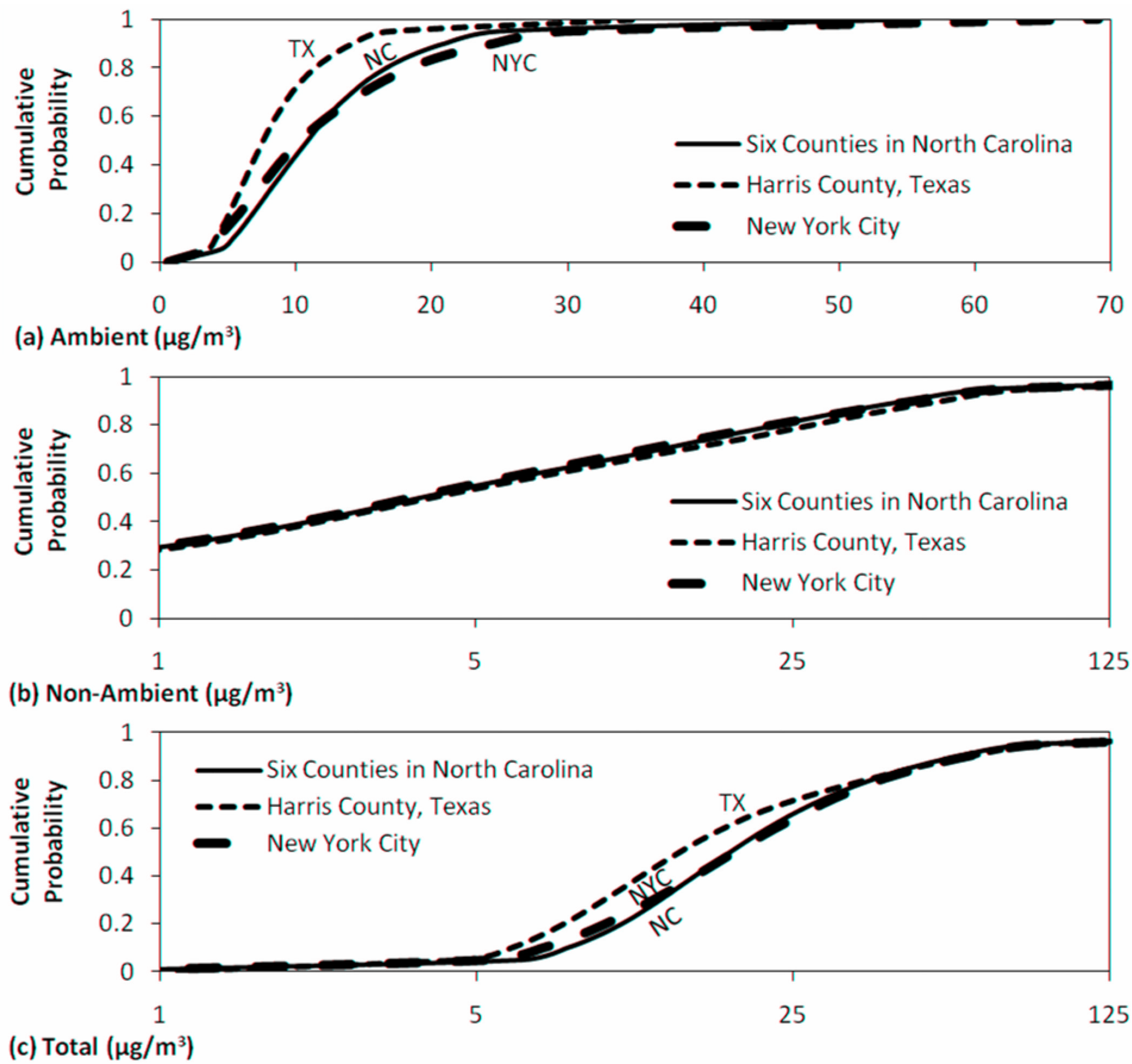

Fig. 2.

Geographic variability of inter-individual variability in daily average $PM_{2.5}$ exposure in summer

Note: in (b), non-ambient exposure is zero below 10th percentile

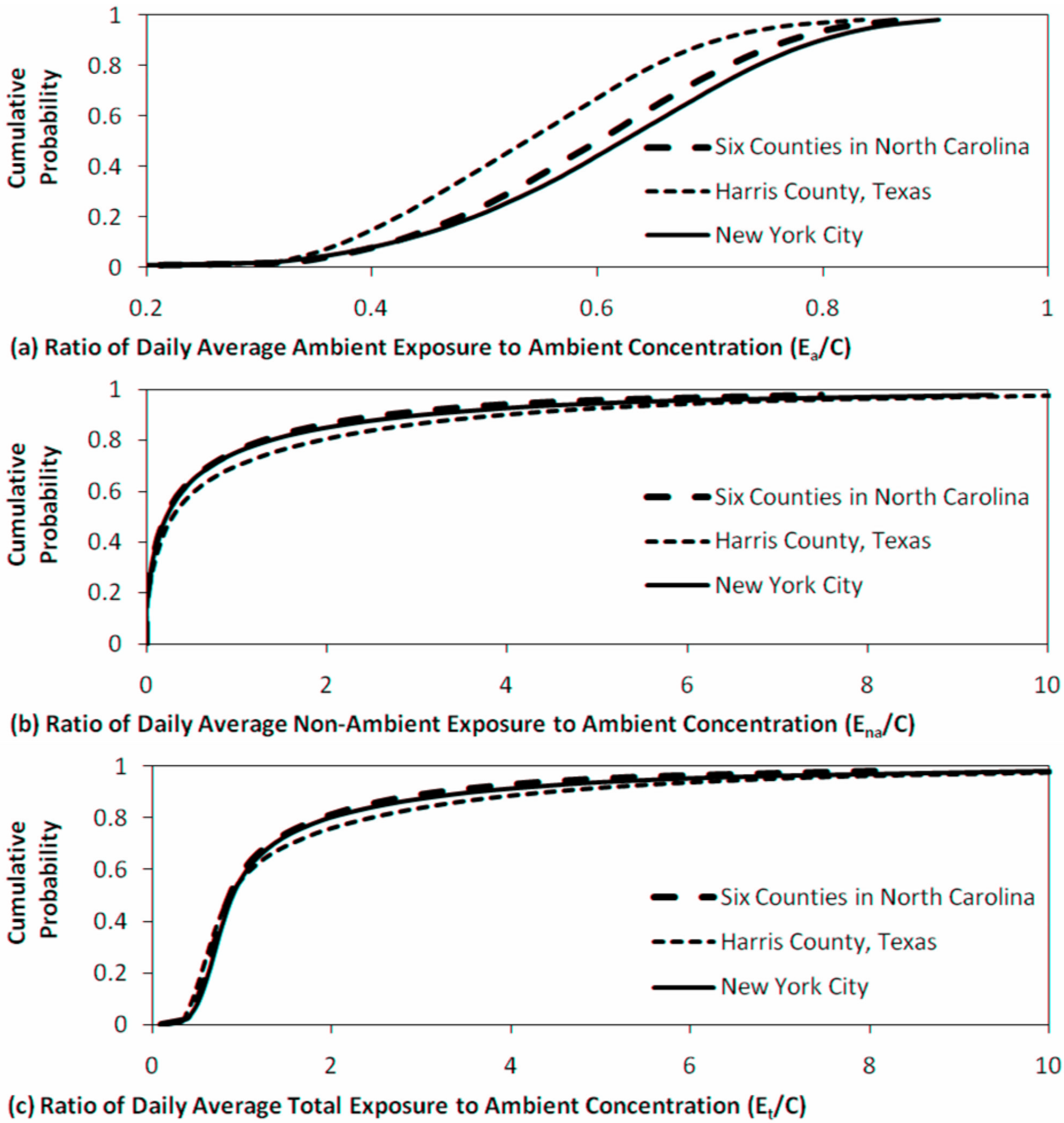

Fig. 3.
Inter-individual variability of exposure to concentration ratio (E/C) in summer

Table 1

Input assumptions for sensitivity analysis of P, k, and ACH

Input	Distribution
P_1	N (0.85, 0.075)[a]
P_2	N (0.90, 0.05)
P_3	Tri (0.70, 0.78, 1.0)[b]
k_1 (h^{-1})	N (0.40, 0.10)
k_2 (h^{-1})	N (0.39, 0.085)
ACH_1 (h^{-1}) in summer in TX[c]	Log (1.05, 2.49)[d]
ACH_2 (h^{-1}) in summer in TX[e]	Log (0.37, 1.90)

[a] N~ Normal (mean, standard deviation).

[b] Tri ~ Triangular (minimum, mode, maximum).

[c] Sources: Murray and Burmaster (1995).

[d] Log ~ lognormal (geometric mean, geometric standard deviation).

[e] Sources: Weisel *et al.,* 2005.

Table 2

Factors affecting geographic variability in daily average $PM_{2.5}$ exposure

Air Exchange Rate for Summer (h^{-1})[a]		
New York City	**Harris County, TX**	**Six Counties Region, NC**
$\mu_g = 0.64\ h^{-1}$, $\sigma_g = 2.09$	$\mu_g = 0.37\ h^{-1}$, $\sigma_g = 1.90$	$\mu_g = 0.54\ h^{-1}$, $\sigma_g = 1.70$

Distribution of Smoking Prevalence by Gender and Age[b] (%)						
	New York City		Harris County, TX		Six County Region, NC	
Age	Male	Female	Male	Female	Male	Female
12–13	4.0	4.0	9.7	6.5	9.4	8.8
14–15	12.0	12.0	10.6	8.0	11.5	12.1
16–17	22.0	18.0	13.5	11.0	20.7	15.1
18–24	28.8	23.4	27.7	20.2	42.3	30.5
25–34	28.5	23.1	27.6	19.1	36.4	20.9
35–44	23.9	18.5	30.0	22.5	29.3	30.5
45–64	24.4	21.7	27.8	20.3	27.7	20.9
>64	12.0	6.6	16.7	9.2	18.5	10.5

Distribution of Population by Gender and Age[c] (%)						
	New York City		Harris County, TX		Six County Region, NC	
Age	Male	Female	Male	Female	Male	Female
12–13	3.6	3.6	3.8	3.9	3.4	3.4
14–15	3.6	3.6	3.7	3.7	3.5	3.5
16–17	3.5	3.6	3.7	3.8	3.6	3.6
18–24	3.3	3.5	3.8	3.7	3.9	3.9
25–34	7.9	8	8.4	8.6	8.3	8.4
35–44	8.8	8.9	9.9	10.2	9.2	9.3
45–64	12.3	12.4	11.3	11.4	11.8	11.9

Air Exchange Rate for Summer (h^{-1})[a]						
New York City		**Harris County, TX**			**Six Counties Region, NC**	
$\mu_g = 0.64\ h^{-1}$, $\sigma_g = 2.09$		**$\mu_g = 0.37\ h^{-1}$, $\sigma_g = 1.90$**			**$\mu_g = 0.54\ h^{-1}$, $\sigma_g = 1.70$**	
>64	6.5	6.9	4.8	5.3	5.9	6.4

Distribution of Housing Types[c] (%)			
Housing Type[d]	New York City	Harris County, TX	Six County Region, NC
Single Family Detached	9.5	68.3	62.5
Single Family Attached	7.2	1.2	4.8
Multiple Family	83.2	8.8	27.2
Mobile Home	0.1	21.7	5.5

[a]Distribution type: lognormal distribution, μ_g: geometric mean, σ_g: geometric standard deviation (Koontz and Rector 1995; Weisel *et al.,* 2005; Wallace *et al.,* 2006).

[b]NCHS (2007): adults 18 years and older; SAMSHA (2008): adolescents 12–17 years old.

[c]U.S. Census 2000. For each geographic area, the percentages shown are for the population 12 and older for both genders, and sum to 100% for all age groups and both genders.

[d]Average Indoor volume: Single Family Detached: 466 m^3; Single Family Attached: 371 m^3; Multiple Family: 241 m^3; Mobile Home: 222 m^3.

NIH-PA Author Manuscript
NIH-PA Author Manuscript

Table 3

Distribution of daily average time spent outdoors and indoors by gender and age[a]

	Time Spent Outdoors Per Day (hr) [b]		Time Spent Indoors Per Day (hr) [c]		Travel Time Per Day (hr) [d]	
Age	Male	Female	Male	Female	Male	Female
12–13	0.9	0.4	19.5	21.1	3.6	2.5
14–15	0.6	0.3	18.4	18.9	5.0	4.8
16–17	0.8	0.2	19.5	18.3	3.7	5.5
18–24	0.8	0.2	20.0	20.6	3.2	3.2
25–34	1.0	0.3	20.5	20.0	2.5	3.7
35–44	1.1	0.2	20.5	20.0	2.4	3.8
45–64	1.0	0.3	21.1	21.1	1.9	2.6
>64	1.0	0.3	21.6	22.7	1.4	1.0

[a]Source: Consolidated Human Activity Database (CHAD) (McCurdy *et al.*, 2005).

[b]Outdoor includes street, parking lot, gas station, park, playgrounds, pool, farm, and all other outdoor microenvironments.

[c]Indoor includes home, office, school, store, bar, restaurant, and all other indoor microenvironments.

[d]Travel includes travel by car, truck, motorcycle, bus, train, subway, airplane, boat, walking, bicycle, and waiting for travel either indoors or outdoors.