Atmospheric Environment 45 (2011) 5684–5691

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

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

Ye Cao [1], H. Christopher Frey*

Department of Civil, Construction and Environmental Engineering, North Carolina State University, Campus Box 7908, Raleigh, NC 27695-7908, United States

ARTICLE INFO

Article history:
Received 8 April 2011
Received in revised form
13 July 2011
Accepted 19 July 2011

Keywords:
Exposure
Particulate matter
Variability
Air exchange rate
Penetration factor
Deposition rate

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 non-ambient 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.

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).

* Corresponding author. Tel.: +1 919 515 1155; fax: +1 919 515 7908.
E-mail addresses: ginnycao@gmail.com (Y. Cao), frey@ncsu.edu (H. C. Frey).
[1] Tel.: +1 919 515 4232; fax: +1 919 515 7908.

1352-2310/$ – see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2011.07.034

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:

$$C_{\text{Home}} = \frac{P \times \text{ACH}}{\text{ACH} + k} C_{\text{ambient}} + \frac{\sum E_i}{(\text{ACH} + k)VT} \quad (1)$$

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.

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, deposition, 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, non-ambient, and total