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Estimation of residential fine particulate matter infiltration in Shanghai, China☆

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ABSTRACT

Ambient concentrations of fine particulate matter ($PM_{2.5}$) concentration is often used as an exposure surrogate to estimate $PM_{2.5}$ health effects in epidemiological studies. Ignoring the potential variations in the amount of outdoor $PM_{2.5}$ infiltrating into indoor environments will cause exposure misclassification, especially when people spend most of their time indoors. As it is not feasible to measure the $PM_{2.5}$ infiltration factor (F_{inf}) for each individual residence, we aimed to build models for residential $PM_{2.5}$ F_{inf} prediction and to evaluate seasonal F_{inf} variations among residences. We repeated collected paired indoor and outdoor $PM_{2.5}$ filter samples for 7 continuous days in each of the three seasons (hot, cold and transitional seasons) from 48 typical homes of Shanghai, China. $PM_{2.5}$-bound sulfur on the filters was measured by X-ray fluorescence for $PM_{2.5}$ F_{inf} calculation. We then used stepwise-multiple linear regression to construct season-specific models with climatic variables and questionnaire-based predictors. All models were evaluated by the coefficient of determination (R^2) and root mean square error (RMSE) from a leave-one-out-cross-validation (LOOCV). The 7-day mean (±SD) of $PM_{2.5}$ F_{inf} across all observations was 0.83 (±0.18). F_{inf} was found higher and more varied in transitional season (12–25 °C) than hot (>25 °C) and cold (<12 °C) seasons. Air conditioning use and meteorological factors were the most important predictors during hot and cold seasons; Floor of residence and building age were the best transitional season predictors. The models predicted 60.0%–68.4% of the variance in 7-day averages of F_{inf}. The LOOCV analysis showed an R^2 of 0.52 and an RMSE of 0.11. Our finding of large variation in residential $PM_{2.5}$ F_{inf} between seasons and across residences within season indicated the important source of outdoor-generated $PM_{2.5}$ exposure heterogeneity in epidemiologic studies. Our models based on readily available data may potentially improve the accuracy of estimates of the health effects of $PM_{2.5}$ exposure.

1. Introduction

Epidemiologic studies have consistently suggested fine particulate matter ($PM_{2.5}$) as a risk factor for adverse health effects (Pope and Dockery, 2006). However, interpretation of findings from these studies have been hampered by uncertainties in exposures, because outdoor concentrations were universally used as an exposure proxy, even though most individuals spend more than 80% of their time indoors (Leech et al., 1996; Klepeis et al., 2001; EPA, 2013). Although it has been found indoor $PM_{2.5}$ commonly correlated with

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ambient concentrations as outdoor $PM_{2.5}$ can enter the indoor spaces (Chen and Zhao, 2011), spatial and temporal variations of $PM_{2.5}$ outdoor-to-indoor transport haven't been fully understood, which is needed for exposure assessment methods improvement.

$PM_{2.5}$ infiltration factor (F_{inf}), defined as the equilibrium proportion of outdoor fine particles that penetrates indoors and remains suspended (Chen and Zhao, 2011), was useful for quantifying the fraction of the total indoor particles with outdoor origin. Studies conducted in North America and Europe found substantially spatial and temporal variation of $PM_{2.5}$ F_{inf} (Chen and Zhao, 2011), which indicates that ignoring potential variations in the outdoor-indoor $PM_{2.5}$ infiltration would result in exposure misclassification (Allen et al., 2007; Meng et al., 2005; Long et al., 2001) that could further bias health effect estimates.

Particle-bound sulfate or sulfur has been commonly used to estimate $PM_{2.5}$ F_{inf} for residential homes (Wallace and Williams, 2005; Dockery and Spengler, 1967; Sarnat et al., 2002), because it is abundant in ambient particles, especially in the submicron particle size range (Hänninen et al., 2004) and with few indoor sources (Sarnat et al., 2002). This method requires both indoor and outdoor pollution measurements. However, it is extremely challenging to measure $PM_{2.5}$ F_{inf} for all individual residences in large population studies and establishing F_{inf} prediction models with available data on housing, environment and activities factors (Clark et al., 2010) could be a feasible solution.

Previous studies showed that $PM_{2.5}$ F_{inf} were differently influenced by residential factors between regions. Hystad et al (2009) used temperature, building value, and heating approaches to predict 54% of infiltration among detached residences from the U.S and Canada (Hystad et al., 2009). Chan et al (2005) found year of construction, size of dwelling and category of dwelling energy efficiency were important predictors of PM infiltration across the U.S. (Chan et al., 2005).

According to these previous studies, predictors of F_{inf} varied with regions and climates and the accessibility of some variables may differ from regions as well. F_{inf} models therefore may not be transferable to other locations. In China, only a limited number of studies have evaluated the variation of residential $PM_{2.5}$ F_{inf} between residences and within residence across seasons (Shi et al., 2015). In addition, even fewer studies explained the variation of $PM_{2.5}$ F_{inf} through modeling methods. In this study, we aim to estimate the infiltration of $PM_{2.5}$ in typical homes of Shanghai, China and to investigate key factors of residential $PM_{2.5}$ F_{inf} by establishing prediction models.

2. Methods

2.1. Study design

In this study, residences from the downtown area of Shanghai were recruited through flyers. To rule out the possibility of significant indoor PM or sulfur sources, such as smoking, frying, grilling and candle burning (Gorjinezhad et al., 2017; Amouei Torkmahalleh et al., 2017), we excluded residences with the following residences: 1) residences with smoking family members; 2) those using coal or wood as cooking fuels; 3) those with open kitchens; 4) those having habits of candle burning.

Among residences that met our criteria, apartment and Shikumen (a traditional Shanghainese architectural style characterized by brick-wood structure houses with shared stone gates and patios with lanes and alleys) were selected since they were typical building types in Shanghai and comprise more than 50% of the total housing stock of the city according to the Shanghai Yellow Pages.

2.2. Data collection

A total of 48 recruited residences were eventually monitored between June 2013 and January 2014. Indoor and outdoor sampling were conducted at participants' homes. For indoor sampling, equipment was set in the middle of the main activity room away from kitchens, air conditioners and ventilation. Outdoor sampling equipment was placed in the back yard, away from all structures; whereas for high-rise apartments the outdoor samplers were extended approximately 1-m out of an available window, with any cracks being sealed to prevent air exchange. At each residential site, measurements were conducted for three 7-day periods representing hot, cold and transitional season. All 48 homes had indoor and outdoor sampling equipment running simultaneously in both transitional and cold seasons. For the hot season, indoor $PM_{2.5}$ of 48 homes were monitored, however, only 19 homes had outdoor sampler due to the limited equipment availability.

We used samplers with a 2 L/min pump (PCXR8, SKC Inc., PA, USA) and a $PM_{2.5}$ impactor for indoor and outdoor sampling. To prevent overloading, effectively 72-h samples were collected on pre-weighed 37 mm Teflon filters (225–8303, SKC Inc., PA, USA) using a programmed schedule for each sampling event. All filters were pre-conditioned for 48 h prior to weighing at a constant air temperature of 20 °C ± 1 °C and constant relative humidity (RH) of 50% ± 5%. Field blanks comprised 10% of the total number of collected samples, and blank-corrected $PM_{2.5}$ mass concentrations were determined following gravimetric analysis. PM-bound sulfur in the filters was analyzed using energy dispersive X-ray fluorescence (Cooper Environmental Services, Portland, OR, USA). Real-time indoor and outdoor temperatures and relative humidity during each sampling period were recorded using data loggers (HOBO U10-003).

Information of resident behaviors related to F_{inf} and residence characteristics were gathered through a main questionnaire at recruitment, including building type and year constructed, family members, presence of air conditioning (AC) and heating facilities, presence of air filters/cleaners and sources of indoor particles (cooking fuel type and habits). For behaviors that vary seasonally or typical activities that may occur occasionally, participants were asked to record them with detailed information in a structured questionnaire during each sampling period, including activities related to ventilation, use of AC/heat, time and frequency of cooking and cleaning, and guest smoking.

2.3. F_{inf} calculation

F_{inf} of $PM_{2.5}$ was calculated based on sulfur infiltration factor. First, we calculated the sulfur infiltration factor using Eq. (1) for each residence based on the assumption that there are typically no indoor sources of sulfur.

$$F_{inf}{}^{S}{}_{i} = C^{S}_{Ii}/C^{S}_{Oi} \tag{1}$$

where i means individual-specific; $F_{inf}{}^{S}{}_{i}$ is PM-sulfur infiltration factor for residence i; C^{S}_{Ii} and C^{S}_{Oi} are the indoor (I) and outdoor (O) concentrations of sulfur for residence i, respectively.

Previous studies have reported that $F_{inf}{}^{PM2.5}$ (F_{inf} of $PM_{2.5}$) may differ from that of PM-bound sulfur possibly due to the change of sulfur proportion on $PM_{2.5}$ during the infiltration (Hänninen et al., 2004). Thus, for each season, the observed difference in $PM_{2.5}$ and sulfur infiltration factors was then corrected using the ratio of the corresponding regression coefficients according to Eq. (2).

$$F_{inf}{}^{PM2.5}{}_{i} = (\beta^{PM2.5}/\beta^{S})_{s} \times F_{inf}{}^{S}{}_{i} \text{ (Hänninen et al., 2004)} \tag{2}$$