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Evaluation and Comparison of Measurement Methods for Personal Exposure to Fine Particles in Beijing, China

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Abstract Ambient measurement and microenvironmental modeling were compared with personal measurement in Beijing, China to evaluate their capacity to determine personal exposure to $PM_{2.5}$. The comparison showed the association was insignificant between ambient and personal concentrations, but was significant between modeled and personal concentrations. The association between ambient and personal concentrations was improved for non-smoking dormitories, on heavily polluted days and on weekdays. The median difference was 41% between ambient and personal concentrations and 17% between modeled and personal concentrations. The factors affecting the association and agreement between methods were indoor sources and ubiquitous "personal cloud".

Keywords Exposure · Measurement · $PM_{2.5}$ · China

Personal exposure to airborne particulate matter (PM) can be determined directly by personal measurement and indirectly by ambient measurement at a centrally located site or by microenvironmental models which estimate personal exposure by integrating PM concentrations in microenvironments over time fractions people spent there (Ott 1982; Lioy 1995; Monn 2001). Because of its availability, the ambient measurement has been widely used in epidemiological studies in China to assess personal exposure and to link the exposure to health effects. The studies indicated an increase of 0.38% in mortality with 10 μg/m^3 increase in ambient PM_{10} (PM with the aerodynamic diameter less than 10 μm) concentration in China (Kan and Chen 2002; Chen et al. 2004). However, there is a concern over the validity of ambient concentration as an indicator of personal exposure, because many Chinese people spend more than 80% of their time indoors (Wu et al. 2003; Cao et al. 1997; Zhao et al. 2009). They may have additional exposure there from Chinese cooking, smoking and incense burning which are typical indoor sources related to Chinese culture and living styles (Lung et al. 2007). Moreover, ambient measurement in China is city/region rather than community based monitoring, and therefore may not well reflect human exposure in communities given considerable spatial variability of PM pollution. In spite of this, the concern did not receive proper attention in China until recently when concept of human exposure and personal measurement methods are gradually introduced. An exposure assessment study published in 2004 showed that ambient concentrations of PM_{10} at government administrated stations tended to misinterpret pedestrians' exposure in Guangzhou (Zhao et al. 2004). This finding implies an urgency of performing a systematic evaluation of measurement methods for human exposure in China, which has not been well addressed before. By doing so, accuracy in exposure measurement and exposure-effect relationship will be greatly improved.

Within the framework of a study on exposure of college students to air toxics, this paper is to evaluate the capacity of ambient measurement and microenvironmental modeling to determine college students' exposure to $PM_{2.5}$ (PM with the aerodynamic diameter less than 2.5 μm) in comparison with personal measurement and to clarify major factors affecting the capacity.

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Materials and Methods

Averaged 24-h exposure of college students to $PM_{2.5}$ was determined by ambient measurement, microenvironmental modeling and personal measurement simultaneously in Changping District of Beijing in summer 2008. The concurrent data were then compared to examine their association and agreement. Changping District is located northwest of Beijing with an area of 1,344 km^2 and a population of 504,000. The major sources of $PM_{2.5}$ in summer are local traffic and long distance transported PM. The subjects in this study were non-smoking college students, but some of whom shared dormitories with smoking roommates. They averagely spent 87% of their time indoors.

Personal or indoor $PM_{2.5}$ sample was collected on a 37-mm quartz fiber filter housed in a SKC personal environmental monitor (PEM) connected to a Leland Legacy pump (SKC Inc., Eighty-Four, Pennsylvania, USA) with a flow rate of 10 L/min for 24 h. Although Teflon filter is recommended by US EPA for $PM_{2.5}$ monitoring, quartz fiber filter was used here to further determine particulate-associated PAHs in samples. The two filter types were reported to provide comparable gravimetric determination of airborne $PM_{2.5}$ (Chartier and Weitz 1998). For personal sampling, the pump was placed in a backpack with the PEM close to the participant's breathing zone by attaching it to the shoulder strap of the backpack. The samplers were placed next to participants' beds when they slept at night. For indoor sampling, the sampler was deployed 1.2–1.4 m above floor in dormitory, classroom and dinning hall where the students spent 84% of their daily time. Ambient concentrations of $PM_{2.5}$ were obtained at Changpingzhen sampling site which is close (1,000–1,200 m west) to where the participants live and is included in the Ambient Air Monitoring Network administrated by Beijing Environmental Monitoring Center.

During the sampling, baseline questionnaire and time-activity questionnaire were distributed to the participants to collect their daily time-activity patterns and exposure-related information. The participants were asked to fill out the questionnaires including questions on their general information (age, gender, height, weight, health condition), exposure to environmental tobacco smoke (ETS), time spent in several microenvironments, housing/dorm characteristics (cleaning activity, time with windows open, number of daily visitors, pets inside, dormitory painting), outdoor emission sources nearby, and so on, after each individual day of personal measurements. The time budgets of the participants, together with indoor and ambient $PM_{2.5}$ concentrations, were used to calculate the students' exposure to $PM_{2.5}$ by microenvironmental modeling.

$PM_{2.5}$ mass concentration was determined based on the filter weight increase after sampling and the total air volume collected. The filter was conditioned at 20 ± 1°C and 35% ± 5% relative humidity (RH) for at least 24 h, and then was weighed in triplicate on Sartorius LA130S-F analytical balance (Sartorius Inc., Goettingen, Germany). The flow rate of the sampler was calibrated by a Dry-Cal meter (SKC Inc., Eighty-Four, Pennsylvania, USA) and remained 10 ± 0.3 L/min during sampling. Field blanks were 32 ± 30 μg for personal samples and 18 ± 12 μg for indoor samples. Detection limits were 6.2 $\mu g/m^3$ and 2.5 $\mu g/m^3$ for personal and indoor measurements, respectively.

The data analysis was conducted using Statistical Analysis System (SAS Institute, Inc., Cary, North Carolina, USA). The distributions of ambient and personal measurements and modeling results were investigated by Q-Q plots and Shapiro–Wilk test. The associations and the differences between ambient and personal concentrations and between modeled and personal concentrations were examined by Spearman rank correlation analysis (PROC CORR SPEARMAN) and by paired *t*-test (PROC UNIVARIATE), respectively. To investigate the influences of exposure to ETS, ambient air pollution and day of week on the association between ambient and personal concentrations, the correlation analysis and the *t*-test were applied to the data stratified by the factors above.

Results and Discussion

The results from different measurement methods are presented in Table 1. It was shown that ambient measurement and microenvironmental modeling tended to underestimate the students' exposure to $PM_{2.5}$ compared to personal measurement.

To investigate the validity of using the ambient $PM_{2.5}$ concentrations at Changpingzhen as a measure of the college students' exposure, the association between ambient and personal measurements (ambient-personal association) was examined (Table 2). With all the observations, the ambient-personal association was insignificant with the Spearman correlation coefficient (Spearman R) of 0.4649 and p-value of 0.11. Therefore, the use of the ambient concentrations to characterize the students' exposure would suffer from the potential for exposure misclassification. However, after the students living in smoking and merely cleaned dormitories were excluded, the Spearman R increased by 54% to 0.7143, suggesting a marginally significant association between ambient and non-smoking students' exposure ($p = 0.07$). The results indicated that environmental tobacco smoke (ETS) and house dust resuspension were responsible for the deterioration of the

Table 1 Exposure of college students to $PM_{2.5}$ from different measurement methods

Measurement method	Exposure to $PM_{2.5}$ (μg/m^3)		
	Median	Mean ± standard deviation	Range
Ambient measurement	73.2	98.8 ± 46.8	45.6–169
Microenvironmental modeling	97.9	104 ± 32.4	64.1–162
Personal measurement	122	163 ± 110	75.0–470

Table 2 Association and differences between ambient and personal $PM_{2.5}$ concentrations

Category	Spearman R	p^a	Difference % (median and range)	p^b
All observations	0.4649	0.11	41 (2–69)	0.03
Non-smoking dorm	0.7143	0.07	23 (2–50)	0.72
Smoking dorm	0.6957	0.12	57 (41–69)	0.01
Compliance days	−0.0714	0.87	46 (2–69)	0.01
Polluted days	0.8721	0.05	29 (11–64)	0.33
Weekdays	0.6347	0.09	35 (2–69)	0.10
Weekends	0.3000	0.62	50 (11–69)	0.10

[a] The p-value is applied to correlation analysis

[b] The p-value is applied to t-test

strength of ambient-personal association in this study. During the sampling periods, the students with ETS exposure problem spent 11–14 h in their dormitories where the average $PM_{2.5}$ concentration was 100 μg/m^3, while spent only 0.7–2.3 h outdoors where the average $PM_{2.5}$ concentration was 110 μg/m^3. Therefore, the indoor sources, ETS, had more impacts on the students' exposure. This is consistent with previous findings. For example, Janssen et al. (1998) found that excluding days with exposure to ETS improved the correlation between outdoor and personal concentrations from the median Pearson's R of 0.50 to 0.71 in 50- to 70-year-old adults in Amsterdam. Tamura et al. (1996) reported that the correlation between ambient and personal levels was good ($R > 0.8$) in reed-mat flooring and non-smoking households in Tokyo. Chinese cooking was another important indoor source resulting in a high average $PM_{2.5}$ concentration of 699 μg/m^3 in dining halls in this study. However, since most of the students spent similar time (∼1 h) in the dining halls every day, the impact of cooking on the ambient-personal association was not assessed in this study. Additionally, the ambient-personal association was affected by ambient air pollution. The air pollution index (API) was used to divide the data into two groups: data collected on compliance days with the API less than 100 and data collected on heavily polluted days with the API more than 100. The ambient-personal association was stronger on heavily polluted days (Spearman $R = 0.8721$, $p = 0.05$) than on compliance days (Spearman $R = -0.0714$, $p = 0.87$). The ambient $PM_{2.5}$ pollution can contribute personal exposure in direct way (exposing individuals outdoors) and indirect way (penetrating into indoor environment). Given 8–11 h with window open in dormitories in summer, a greater impact of ambient pollution on personal exposure could be expected on heavily polluted days (average ambient $PM_{2.5}$ concentration of 153 μg/m^3) than on compliance days (average ambient $PM_{2.5}$ concentration of 64.8 μg/m^3). The Spearman R was higher on weekdays (0.6347) than on weekends (0.3000). It would be explained by the fact that ambient $PM_{2.5}$ concentrations were higher on weekdays (an average of 115 μg/m^3) than those on weekends (an average of 73.2 μg/m^3), which was also observed at a hot spot for air pollution in New Jersey (Fan et al. 2006).

The differences between ambient and personal measurements (ambient-personal differences) were significant ($p = 0.03$), ranging from 2% to 69% with the median of 41% (Table 2). This could be expected as human exposure to air pollutants is contributed not only by ambient air pollution but also by indoor air pollution and even "personal cloud" (Wu et al. 2005). In non-smoking dormitories, on heavily polluted days and on weekdays, the ambient-personal differences were insignificant ($p = 0.72$, 0.33 and 0.10) and lower (the median difference of 23%, 29% and 35%) than their opposites (the median difference of 57%, 46% and 50%), suggesting outdoor air pollution dominated personal exposure.

Microenvironmental modeling was compared with personal measurement in Table 3. The association between modeled and personally measured concentrations of $PM_{2.5}$ (modeled-personal association) was significant with the

Table 3 Association and differences between modeled and personal $PM_{2.5}$ concentrations

Category	Spearman R	p^a	Difference % (median and range)	p^b
Simultaneous monitoring	0.7857	0.02	17 (2–72)	0.04
Average concentrations	0.7362	0.03	14 (1–76)	0.07
Average time intervals	0.3898	0.53	18 (3–73)	0.04

[a] The p-value is applied to correlation analysis

[b] The p-value is applied to t-test

Spearman R of 0.7857 and p-value of 0.02, which was stronger than the ambient-personal association ($R = 0.4649$, $p = 0.11$) discussed above. The differences between modeled and personally measured concentrations (modeled-personal differences) ranged from 2% to 72% with the median of 17%, which were also lower than the ambient-personal differences (41%, 2%–69%). This suggested that the modeling gave more accurate estimation of the students' exposure than the ambient measurement by taking into account $PM_{2.5}$ pollution in all the locations where the students spent their time. In spite of the improvements, the modeled results still significantly underestimated the exposure ($p = 0.04$ in Table 3). It would indicate the importance of so-called "personal cloud" to personal exposure (Wu et al. 2005), which could not be estimated in the microenvironmental modeling.

A microenvironmental model estimates personal exposure based on measurements in microenvironments (MEs) and time-activity data of population. Theoretically, air monitoring and time-activity survey should be conducted for individuals simultaneously. However, it is demanding and expensive to obtain such refined data in large-scale epidemiological studies. The most feasible approach is to use average concentration in each "group-MEs" and average time budget of each sub-population group (Wu et al. 2003; Cao et al. 1997; Borrego et al. 2006). This alternative approach was assessed in this study (Table 3). Using the average $PM_{2.5}$ concentrations in MEs (dinning hall, classroom, dormitory and outdoors) did not substantially change the modeled-personal association and differences, with the Spearman R and the median difference of 0.7362 and 14% compared with 0.7857 and 17% in use of simultaneous measurements. This could be explained by two reasons. First, in dining halls and classrooms, the day-to-day variation of $PM_{2.5}$ concentrations was low during the sampling periods. The average concentrations could represent the air pollution there. Secondly, although the ambient $PM_{2.5}$ concentrations varied greatly day-to-day, the contribution of ambient air pollution to personal exposure was low (an average of 7%).

The time intervals which the students spent in each ME were averaged, and then students' exposure was re-calculated based on the averaged time intervals (Table 3). As shown in Table 3, the exposure estimated using the average time intervals was poorly associated with the exposure measured ($R = 0.3898$, $p = 0.53$). It would indicate that the modeling could not identify high risk population exposed to high levels of $PM_{2.5}$, because the average time intervals smoothed out the variation of time which different students spent in each ME. However, the use of the average time intervals did not result in larger bias from the measured personal exposure (the median difference of 18%) than the use of the specific time-activity records for individuals (the median difference of 17%).

In conclusion, the findings in this study provided support for using ambient measurements as a measure of exposure to $PM_{2.5}$ for college students living in non-smoking dormitories and on heavily polluted days. Microenvironmental modeling would be recommended because the modeled-personal association was good and the MEs can be well defined for college students and be easily accessed for air morning. This study is a small-scale study designed for college students. To further evaluate the performance of measurement methods for personal exposure and to better understand the association between personal, indoor and outdoor pollution in China, more studies involving other groups of population are needed.

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