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Agricultural burning smoke in Eastern Washington: Part II. Exposure assessment

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Abstract

Several studies have documented potential health effects due to agricultural burning smoke. However, there is a paucity of literature characterizing community residents' exposure to agricultural burning smoke. This study assesses personal exposures to particulate matter (PM) with aerodynamic diameters $<2.5\,\mu m$ ($PM_{2.5}$) from agriculture burning smoke (E_b) for 33 asthmatic adults in Pullman, WA. $PM_{2.5}$ concentrations were measured on 16 subjects, inside of all but four residences, outside of 6 residences, and at a central site. The mean ± standard deviation of personal exposure to $PM_{2.5}$ was $13.8 \pm 11.1\,\mu g\,m^{-3}$, which was on average $8.0\,\mu g\,m^{-3}$ higher during the agricultural burning episodes ($19.0 \pm 11.8\,\mu g\,m^{-3}$) than non-episodes ($11.0 \pm 9.7\,\mu g\,m^{-3}$). The levoglucosan (LG, a unique marker for biomass burning PM) on personal filter samples also was higher during the episodes than non-episodes (0.026 ± 0.030 vs. $0.010 \pm 0.012\,\mu g\,m^{-3}$). We applied the random component superposition model on central-site and home indoor PM measurements, and estimated a central-site infiltration factor between 0.21 and 2.05 for residences with good modeling performance. We combined the source apportionment and total exposure modeling results to estimate individual E_b, which ranged from 1.2 to $6.7\,\mu g\,m^{-3}$ and correlated with personal LG with an r of 0.51. The sensitivity analysis of applying the infiltration efficiency estimated from the recursive model showed that the E_b (range: 1.3–$4.3\,\mu g\,m^{-3}$) obtained from this approach have a higher correlation with personal LG ($r = 0.75$). Nevertheless, the small sample size of personal LG measurements prevents a comparative and conclusive assessment of the model performance. We found a significant between-subject variation between episodes and non-episodes in both the E_b estimates and subjects' activity patterns. This suggests that the LG measurements at the central site may not always represent individual exposures to agricultural burning smoke. We recommend collecting more microenvironmental samples to model the E_b and more personal samples to validate the E_b estimates.

Keywords: Biomass burning; Smoke impact; Personal exposure; Random component superposition model; Recursive model; Spatial variation

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1. Introduction

Agricultural burning (i.e. burning field stubble or waste rice straw) was deemed as a cost-effective way of cleaning and preparing the field for the succeeding growth season (Mazzola et al., 1997). However, smoke from Ag burning contains various air pollutants, including particulate matter (PM), nitrogen dioxide, carbon monoxide, and a series of semi-volatile and volatile organic compounds (Bouble, 1969; Jenkins et al., 1996; Jacobs et al., 1997) that are known to cause adverse health effects. However, the exposure and health effects have been difficult to assess due to the following factors. Agriculture burning typically occurs in rural locations where there is often a limited number of ambient air quality monitors, such that air pollution excursions may not be detected. When the excursions are recorded, these short-term spikes usually do not result in a violation of the annual or 24-h NAAQS for $PM_{2.5}$ and PM_{10}. The sparse population also made epidemiologic studies difficult. Thus, a handful of studies assessing health effects of agriculture burning smoke (Jacobs et al., 1997; Roberts and Corkill, 1998; Torigoe et al., 2000) suffered several limitations. They often relied on the air quality data collected at centrally located monitoring sites, which may not represent the community residents' actual exposures (Ebelt et al., 2000; Liu et al., 2003). These studies also could not directly link the health effects to smoke from agricultural burning. We know of no existing publications that characterize community residents' exposure to Ag burning smoke.

This paper assesses personal exposures to $PM_{2.5}$ from ambient (outdoor) sources (E_a) and agriculture burning smoke (E_b), respectively, during the burning season in Pullman, WA. Our first paper in series (Jimenez et al., 2006) characterized the air quality in Ag burning smoke and estimated a 35% contribution from biomass burning to $PM_{2.5}$. In this paper, we apply a random component superposition (RCS) model (Ott et al., 2000), a recursive mass balance model (Allen et al., 2003; Allen et al., 2004), and a total exposure model (Wilson et al., 2000; Williams et al., 2003; Allen et al., 2004; Wu et al., 2005) to estimate the E_b using subjects' time–place–activity information and $PM_{2.5}$ measurements collected at the central and home indoor sites. In the final paper (Sullivan et al., 2006), we examine the association between health effects and exposure to agricultural burning smoke for these 33 asthmatic adult subjects.

2. Methods

2.1. Study design

This study was conducted in Pullman, WA, from 3 September 2002 to 1 November 2002. The fall burning season usually involved more acreage burned and higher short-term $PM_{2.5}$ concentrations than those observed in the spring (Jimenez, 2002). Pullman is located in eastern WA (population ~25,000), approximately 80 miles south from Spokane, WA. Thirty-three adult subjects with asthma (mean age = 27, min = 18, max = 52) were recruited in this study. These subjects were either students or staff at the Washington State University (WSU), and were typically on the WSU campus during the day. A central site on WSU campus was set up on top of the Dana Hall (~12 m above street level) with various air quality monitors. Continuous $PM_{2.5}$ mass concentrations were measured using a Tapered Element Oscillating Microbalance (TEOM, Series 1400a, Rupprecht & Patashnick Co., Inc). Light scattering was measured using a nephelometer (Radiance Research, Seattle, WA) and integrated 12-hr (starting at 8 AM and 8 PM) $PM_{2.5}$ samples were collected from two collocated Harvard Impactors ($HI_{2.5}$) (Air Diagnostics and Engineering, Inc., Naples, ME). The $HI_{2.5}$ were equipped with Teflon filters for gravimetric and XRF analysis. Some of the Teflon filters were extracted and analyzed with GC/MS for levoglucosan (LG), a tracer for biomass burning smoke (Simpson et al., 2004). The LG detection limit is $0.02\,\mu g\,ml^{-1}$ and the precision is 24%. In addition to our central-site, the Washington State Department of Ecology (WDOE) also monitored the $PM_{2.5}$ routinely with a Radiance Research nephelometer in Pullman (DOE site, Fig. 1). Detailed descriptions about these monitoring methods were provided in Jimenez et al. (2006).

Indoor PM was monitored at 13 subjects' residences during the first 30 days and the rest of the subjects during the second 30 days (Table 1), using either the Radiance Research nephelometer or the personal DataRAM (pDR, Thermo-Andersen, Smyrna, GA). Four subjects did not have indoor PM measurements due to the lack of available instruments. The precision of the nephelometer is 3–8%, and for the pDR it is 12% under $10\,\mu g\,m^{-3}$

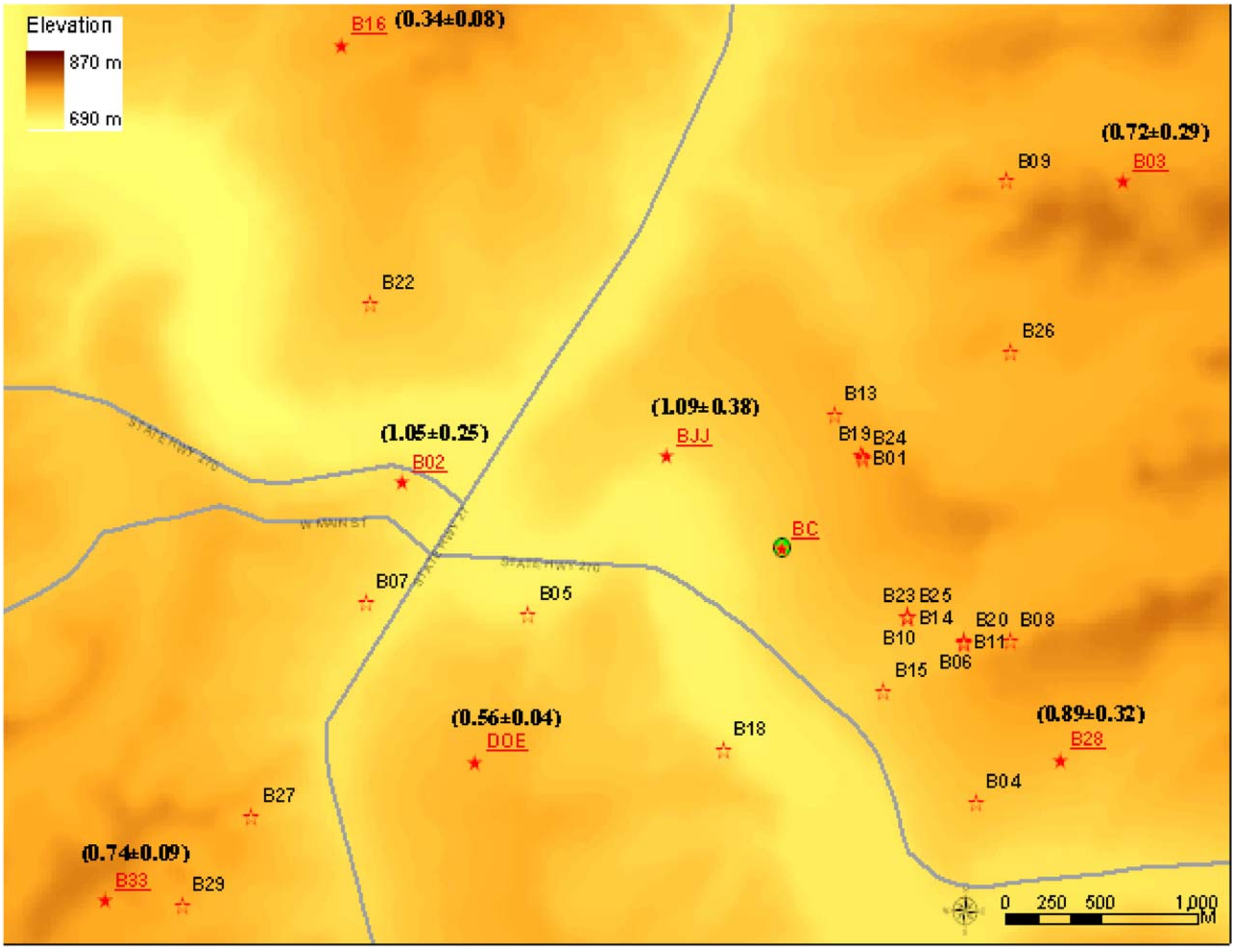

Fig. 1. The locations of the subjects' residences. The underlined ID labels indicate sites with outdoor $PM_{2.5}$ measurements. The mean ratio and the standard deviation of the outdoor to central-site $PM_{2.5}$ were in parenthesis.

Table 1
Numbers of samples collected on subjects, at residences and at the central site by sessions

Location	Instruments	Parameter to the right	Quantity	
			First session	Second session
Personal	HPEM (Teflon)	Subjects/total filters	8[a]/34	8/40
	HPEM (Quartz)	Subjects/total filters	8[a]/34	8/40
Indoor home	Nephelometer	Sites/total hours	9[b]/2795	9[c]/3825
	pDR	Sites/total hours	10[b]/5923	16[c]/6443
Outdoor home	HPEM	Sites/total filters	–	5/32
Central site	Nephelometer	Hours	710	671
	HI (Teflon)	Total filters	60	58

[a]One subject dropped out after monitored for 1 day.
[b]A total of 13 sites were monitored; 6 sites had both a Nephelometer and pDR.
[c]A total of 16 sites were monitored; 9 sites had both a Nephelometer and pDR.

and approximately 5% above 10 $\mu g\,m^{-3}$ (Liu et al., 2002). Each week, two subjects volunteered for personal exposure monitoring for 4 (in the first week) or 5 (in the rest 7 weeks) consecutive days, starting from late afternoon on Monday (Table 1). Each subject kept a time–activity diary (TAD) with a 10-min resolution. Nominal 24-h personal $PM_{2.5}$ samples were collected using two collocated Harvard Personal Environmental Monitors ($HPEM_{2.5}$, Harvard School of Public Health, Boston, MA), each connected to its own personal pump (BGI AFC 400S, Waltham, MA) operated at 4 LPM. The precision ($2.2\,\mu g\,m^{-3}$) and accuracy ($0.4\,\mu g\,m^{-3}$) of the $HPEM_{2.5}$ have been determined in our previous study (Liu et al., 2003). One of the HPEMs contained a Teflon filter for gravimetric analysis and the other contained a quartz filter for EC/OC analysis (Pang et al., 2002; NIOSH, 2003). The Teflon filters were further analyzed for XRF ($N = 30$) and/or LG ($N = 48$).

After a spatial pattern was observed between the central and the DOE sites during the first half of the study period, outdoor $HPEM_{2.5}$ measurements were added at 5 home sites for further characterization of the spatial variation of $PM_{2.5}$ (Table 1). These sites were selected to represent the four hills that were separated by two major cross roads in the valleys (Fig. 1). Each home outdoor sample was collected over approximately 25-h intervals during two Ag burning "episodes" (October 9–11 and 17–19) and over approximately 1 week (144–170 h) otherwise. In addition to the filter samples, continuous $PM_{2.5}$ was also measured by a Radiance Research nephelometer at one of the subjects' residences in northwest Pullman (B16 in Fig. 1). Agricultural burning episodes were declared based on the combination of the central-site TEOM measurements, visual observations, and the regional burn calls from the WDOE and the Idaho Department of Environmental Quality (Jimenez et al., 2006). There were four real episodes, among which two were declared during the monitoring campaign and two were identified later during the data analysis phase. One "sham" episode, i.e. the subjects were informed that there was an episode when there was not, was declared during the monitoring campaign (Jimenez et al., 2006) and was used as a control in health assessment.

2.2. Data analysis

Data analyses were conducted to characterize $PM_{2.5}$ spatial variation and personal exposure. The general linear model (GLM) was used to test the between-subject effects on mass concentration, LG, and EC/OC measurements. When there was no significant between-subject effect, subjects were pooled and the differences in PM exposures during agricultural burning episodes and non-episodes were tested using GLM. A spatial regression model was constructed to examine factors affecting the $PM_{2.5}$ concentrations at the home sites. The dependent variable included measurements from the home outdoor, B16, and the DOE sites with averaging times for nephelometers matching those of the HPEM measurements. The predictors included the matched central-site nephelometer measurements ($\mu g\,m^{-3}$), distance from outdoor sites to the central site (km), elevation (binary variable: below or above the central site), and temperature at the central site (°F). E_a was estimated using the RCS model (Ott et al., 2000; Williams et al., 2003) and total exposure model (Allen et al., 2004; Wu et al., 2005). E_b was estimated by proportioning E_a to agricultural burning related $PM_{2.5}$ based on the chemical mass balance (CMB) modeling results for each 12-h monitoring period (Jimenez et al., 2006).

Estimate the particle infiltration efficiency (F_{inf}): The typical RCS model estimates F_{inf} by regressing home outdoor $PM_{2.5}$ (C_o) against indoor $PM_{2.5}$ (C_i):

$$C_i = b_0 + b_1 C_o, \tag{1}$$

where b_1 represents F_{inf} from the immediate home outdoor to indoor environments and b_0 represents indoor sources and the measurement error. Since home outdoor measurements were not available for most homes in this study, we replaced the C_o in Eq. (1) with the 12-h $HI_{2.5}$ measurements at the central site (C_c), while C_i was the corresponding indoor $PM_{2.5}$ measurements averaging over the same 12-h period. Although the RCS model was originally developed for a large number of homes in a single geographic area (Ott et al., 2000), we extended the model to multiple measurements on a relatively small number of homes. In light of the outlier effects in the RCS model estimates with a small sample size, we applied the Robust regression algorithm (Proc Robustreg in SAS v9.0, SAS Institute Inc., Cary, NC, USA) to Eq. (1) to obtain more stable b_1 estimates. Our b_1 estimate is a modified F_{inf} or a central-site infiltration factor (A), which represents a combined effect of $PM_{2.5}$ spatial variations between the home and the central sites and F_{inf} from home outdoor to home indoor environments, and could be greater than 1. It was calculated based on 14–61 (mean = 44) paired 12-h $PM_{2.5}$ observations for each home.

Estimate personal exposures to ambient-generated PM (E_a): The total exposure model was used to calculate E_a on a 12-h basis:

$$E_a = \alpha_{RCS} C_c = [f_i A + (1 - f_i)]C_c, \quad (2)$$

where α_{RCS} is the ambient contribution fraction (Allen et al., 2004) and f_i is the time fraction spent indoors at home. Because during the school day all subjects were on the WSU campus where the central-site was located, thus minimizing the spatial effects on A, we made a strong assumption that A at school indoors or other indoor microenvironments was 1. As our main interest was to estimate E_b and most of the PM excursions from agricultural burning occurred during the nighttime when subjects were asleep at home (Jimenez et al., 2006), it was not as critical to obtain an accurate A for the school and other indoor microenvironments during the day.

Estimate personal exposures to agricultural-burning PM (E_b). The E_b at any 12-h sampling period was calculated as

$$E_b = (\mathrm{CMB_{VEG}}/C_c)E_a, \quad (3)$$

where $\mathrm{CMB_{VEG}}$ is the 12-h average vegetative burning mass concentration at the central site apportioned by the CMB model (Jimenez et al., 2006). The accuracy of the E_a estimates was evaluated by comparing them with those estimated from the sulfur tracer method (Allen et al., 2003), while E_b was evaluated by comparisons with the LG tracer method.

Sensitivity analyses: The presumption of $A = 1$ at school indoors or other indoor microenvironments was tested by replacing A at these two microenvironments with varying values in Eq. (2) and evaluating the effects on the E_b estimates. To test the robustness of the A and E_b estimates, we also estimated F_{inf} directly using a recursive model (RM) (Allen et al., 2003; Wu et al., 2005):

$$(b_{sp})_t^{in} = a_1(b_{sp})_t^{out} + a_2(b_{sp})_{t-1}^{in} + S_t^{in}, \quad (4)$$

where $(b_{sp})_t^{in}$ is the indoor light scattering value at time t, i.e. hourly indoor $PM_{2.5}$ at residence measured with either nephelometer or pDR; $(b_{sp})_t^{out}$ is the outdoor light scattering value at time t, i.e. hourly outdoor $PM_{2.5}$ at residence estimated from the GLM spatial model; $(b_{sp})_{t-1}^{in}$ is the indoor light scattering value at a previous time $t-1$; and S_t^{in} is the contribution from indoor sources. The a_1 and a_2 in Eq. (4) were obtained through linear regression with the effect of S_t^{in} minimized by censoring the data for indoor sources. F_{inf} was then calculated as $a_1/(1-a_2)$. Both the RCS model and RM rely on certain correlations between the central-site and the home $PM_{2.5}$ measurements. Site B32 was excluded from both models as it was located outside the main Pullman area. More detailed RM approach can be found in Appendix A. E_a was estimated as

$$E_a = (f_i F_{inf} + f_o)C_o + (1 - f_i - f_o)C_c, \quad (5)$$

where f_o is the time fraction spent at home outdoors and C_o is the outdoor $PM_{2.5}$ at residence estimated from the GLM spatial model. The sum of the coefficients for C_o and C_c in Eq. (5) is the ambient contribution fraction (α_{RM}). E_b was recalculated according to Eq. (3).

3. Results and discussion

3.1. Quality control

A total of 118 $HI_{2.5}$ samples with Teflon filters were collected at the central site, and 74 and 44 $HPEM_{2.5}$ samples were collected on the subjects and outside the subjects' home, respectively. Five $HI_{2.5}$ samples with Teflon filters at the central site were removed from the dataset due to either filter (e.g. filter mishandled or damaged) or air flow problems (e.g. not within 10% of the designated flow rate). Five personal Teflon, 9 personal quartz and 5 home outdoor samples were removed due to similar fatal errors. All filter samples were blank corrected (mean ± standard deviation: 1.5 ± 4.1 and 7.7 ± 5.2 μg for $HI_{2.5}$ and $HPEM_{2.5}$ samples, respectively). The nephelometer measurements at the central site were converted to mass concentrations based on the calibration against the collocated $HI_{2.5}$ measurements. The pDRs were collocated with and calibrated against the central-site nephelometer over 4 days at the end of the study period. The calibration curve for the pDRs had an R^2 ranging between 0.986 and 0.992. The histogram of LG revealed that there was one potential outlier (0.17 μg m^{-3}) with a confirmation from the TAD that this subject was exposed to smoke from a woodstove fire for 160 min. Since we used LG as a tracer for agricultural burning related smoke, this sample was removed from the data set.

3.2. $PM_{2.5}$ spatial analysis

During the second-half of the study, the average $PM_{2.5}$ concentrations at the central site and other outdoor (home outdoor/DOE) sites were 14.8 ± 4.6 and 11.7 ± 6.1 μg m^{-3}, respectively. The average elevation of these other outdoor sites was 757 ± 19 m (range: 732–790 m) with 3 sites located above the central site, and the average distance from other outdoor to the central site was 1.2 ± 0.6 km (range: 0.4–2.1 km). The average temperature was 42.4 ± 6.7 °F (range: 28.8–51.9 °F). $PM_{2.5}$ concentrations were generally higher in the valley and decreased with elevations. The ratios of home outdoor to central site $PM_{2.5}$ measurements ranged between 0.27 and 1.81, with the lowest mean ratios observed for the hill site B16 (mean ± ⊖ SD = 0.34 ± 0.08), median for B33 (0.74 ± 0.09), and the highest for valley site BJJ (1.09 ± 0.38) (Fig. 1). Four sites (i.e. B02, B03, B28, and BJJ) had a relatively larger variation (SD > 0.2) in the home outdoor/central site ratio over the monitoring period while the other three sites had small variation (SD < 0.2), suggesting a possible temporal–spatial interaction among these sites. Site B16, located on the northwest hill facing away from the central Pullman valley, was likely in a different airshed as it had the lowest ratio and the lowest correlation with the central site measurements ($r = 0.25$). The observed vertical concentration gradient was probably due to the nighttime drainage flow (or mountain wind) that brought upper-layer PM remaining from the daytime agricultural burning to the ground level. The vertical gradient could also be a result of the residential wood burning. However, based on the 2000 census data there were only 30 (0.3%) housing units in Pullman using wood as the heating source.

Significant predicting factors for the spatial variation included the central site PM measurements, home elevation, and outdoor temperature ($R^2 = 0.63$ without B16 and 0.49 with B16) (Table 2). As none of the other home sites were located near B16 (Fig. 1), B16 was excluded in the final spatial model. The distance from the home to the central site was the least and insignificant contributor to the total R^2 (partial $R^2 < 0.01$, $p = 0.81$). This is partially due to the high correlation ($r = 0.88$) between elevation and distance. Outdoor $PM_{2.5}$ at sites above the central site was on average 4.7 μg m^{-3} lower than those below the central sites. Outdoor $PM_{2.5}$ was positively associated with temperature. Vukovich and Sherwell (2002) also found positive correlation between $PM_{2.5}$ and temperature at an urban site (Washington, DC) and a remote site (Shenandoah National Park, VA) based on principal component analysis.

3.3. Characterization of personal exposure

Subjects spent an average of 19% of their time at school or work and 61% of their time at home indoors (Table 3). To test the Hawthorne effect (Franke and Kaul, 1978), i.e., how subjects might change their activities simply due to the participation in the study or the declaration of episodes, we examined the overall time spent in each microenvironment during declared episode and non-episode days. Using a GLM, we found significant differences in time spent at the home indoors, in transit, and indoors in other microenvironments (Table 3). During declared episodes (i.e. real and sham), subjects spent less time indoors at home and more time in transit or indoors away from home than during non-declared episode periods. The differences remained even when limited to weekdays only. We attributed these differences partially to the fact that subjects had to come to our lab for respiratory health tests during episodes, thus increasing the time spent in transit or indoors away from home. Because of the observed Hawthorne effect, the time–activity pattern and personal PM exposure levels found in this study might not be truly representative of the 'normal' condition. We also

Table 2
The spatial modeling results. The dependent variable is the HPEM samples at home outdoor locations and nephelometer data at the DOE site ($N = 37$)

Parameter		GLM				Stepwise regression			
		Estimate	SE	t value	Pr > |t|	Partial R^2	Model R^2	F value	Pr > F
Intercept		−6.46	4.72	−1.37	0.18				
Neph $PM_{2.5}$ (μg m^{-3})		0.89	0.14	6.59	<.001	0.43	0.43	26.04	<.001
Elevation	Above central site	−4.74	1.26	−3.78	<.001	0.16	0.59	13.02	<.001
	Below central site	0.00	—	—	—				
Temperature at the central site (°F)		0.19	0.10	2.00	0.05	0.04	0.63	4.00	0.05

Table 3
Percent of time spent in each microenvironment

Microenvironment	All	Declared as episode[a]		Real episode[b]	
	Mean (SD) $N=1419$	No Mean (SD) $N=1147$	Yes Mean (SD) $N=272$	No Mean(SD) $N=1071$	Yes Mean(SD) $N=348$
Indoors at home	61.1(20.6)	62.4(19.8)**	55.3(22.7)	62.1(19.9)**	57.9(22.1)
Outdoors near home	0.5(2.3)	0.5(2.3)	0.5(2.0)	0.5(2.4)	0.5(1.9)
Transportation	6.5(5.6)	6.2(5.6)**	8.0(5.4)	6.3(5.7)*	7.2(5.3)
Work	18.9(15.8)	18.8(15.6)	19.4(16.7)	19.0(15.6)	18.7(16.3)
Outdoors away home	2.5(6.5)	2.3(6.2)	3.1(7.8)	2.4(6.3)	2.9(7.2)
Indoors away home	10.5(17.7)	9.7(17.0)**	13.6(20.3)	9.7(16.9)**	12.8(19.9)

**Significantly different between episodes and non-episodes, $p<0.01$.
*Significantly different between episodes and non-episodes, $p<0.05$.
[a]The two episodes declared during the field campaign plus the 'sham' episode.
[b]The two episodes declared during the field campaign plus the two episodes identified during the data analysis phase.

examined whether subjects have different activities during *real* episode vs. non-episode days (regardless of episode declaration) and obtained similar results as to those analyzed by declaration status (Table 3).

In addition, results from the GLM with subject ID and the status of real episode as the fixed effects showed that there was a significant subject effect in time fractions spent in these microenvironments. This between-subject variation on activity patterns might bias the health effect assessment if the analysis was performed based on binary coding of episode status or using the central-site measurements to represent individual exposures. Applying personal exposure measurements or estimates that incorporated the subjects' activity patterns can effectively avoid such a bias.

The mean personal exposure to $PM_{2.5}$ among the 16 monitored subjects was $13.8 \pm 11.1\,\mu g\,m^{-3}$ ($N=68$, median $= 11.4\,\mu g\,m^{-3}$). Results from GLM showed that there was no significant difference in PM exposure among subjects ($p = 0.12$). Thus we pooled all subjects to test for episode effects. The personal exposure was on average $8.0\,\mu g\,m^{-3}$ higher during the episodes ($N = 24$, mean $= 19.0 \pm 11.8\,\mu g\,m^{-3}$) than non-episodes ($N = 44$, mean $= 11.0 \pm 9.7\,\mu g\,m^{-3}$). The mean personal exposure to EC was $0.4 \pm 0.5\,\mu g\,m^{-3}$ and $8.5 \pm 2.7\,\mu g\,m^{-3}$ to OC ($N = 64$). Personal OC exposures were higher during episodes than during other times (10.2 ± 2.8 vs. $7.7 \pm 2.2\,\mu g\,m^{-3}$). However, there was also a significant subject effect ($p<0.01$) on OC exposure. Due to the small sample size, we could not separate the episode effect from the subject effect.

LG in PM samples has been used previously in source apportionment analyses as a unique tracer for biomass combustion (Simpson et al., 2004; Zheng et al., 2002; Larson et al., 2004; Jimenez et al., 2006). The mean personal LG exposure in our study was $0.018 \pm 0.024\,\mu g\,m^{-3}$ ($N = 47$, median $= 0.012\,\mu g\,m^{-3}$) after correcting for the analytical recovery (mean $= 82 \pm 8\%$). This level is lower than those measured at our central site (mean-$0.074\,\mu g\,m^{-3}$) and in southeastern US ($0.166\,\mu g\,m^{-3}$, Zheng et al., 2002) and Seattle, WA ($0.204\,\mu g\,m^{-3}$, Larson et al., 2004). Personal LG exposures were higher during episodes than during other times (0.026 ± 0.030 vs. $0.010 \pm 0.012\,\mu g\,m^{-3}$). Similar to the personal OC exposures, we could not separate the episode effect from the subject effect on the LG measurements ($p<0.01$).

The cross-sectional Pearson correlation coefficient between the central-site and personal $PM_{2.5}$ measurements was 0.29, identical to the 0.29 reported in Seattle, WA (Liu et al., 2003) and higher than the 0.15 in Vancouver, BC (Ebelt et al., 2000). From the scatter plot, 5 personal samples ($>22\,\mu g\,m^{-3}$, denoted as open circles in Fig. 2) were identified as heavily influenced by indoor sources or personal activities. Four of these five samples occurred when the subjects spent more than 2 h at social events during the corresponding sampling period, while the 5th sample occurred when the subject conducted 5 h of unspecified activities. After removing these 5 observations, the cross-sectional correlation increased to 0.65 (Fig. 2), similar to the 0.67 in Baltimore, MD (Sarnat et al., 2000). Previous studies showed that the median

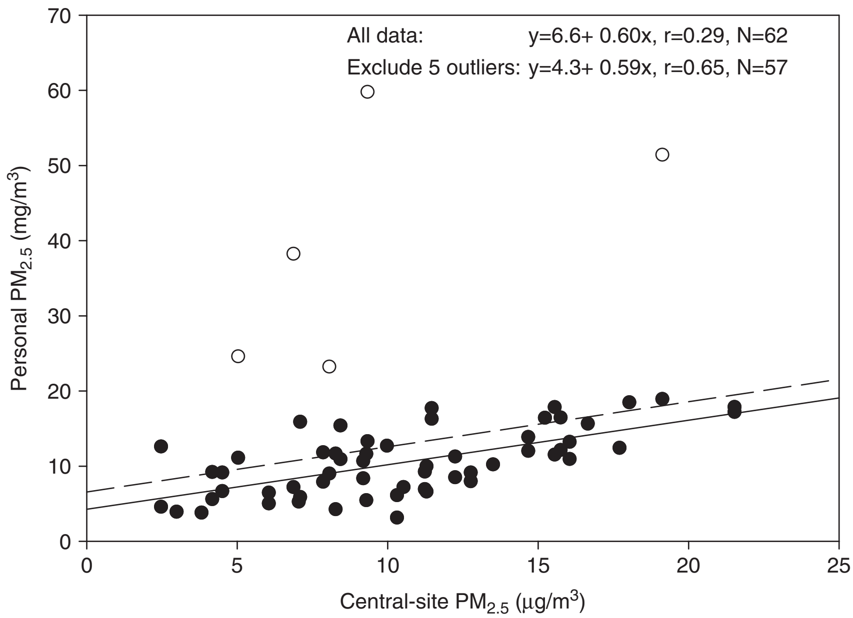

Fig. 2. Correlation between personal and central-site $PM_{2.5}$ measurements. The blank circle represents the five potential outliers.

longitudinal correlation usually was higher than the cross-sectional correlation because the interpersonal variability was eliminated in the longitudinal correlation (Ebelt et al., 2000; Sarnat et al., 2000; Wallace, 2000; Liu et al., 2003). In this study, the longitudinal Pearson correlation coefficient between central site and personal $PM_{2.5}$ for individual subjects ($N = 11$) with more than 3 daily observations ranged between -0.67 and 0.98, with a median of 0.52. The negative longitudinal correlations are likely due to the limited number of samples per subject (N ranged from 3 to 5) (Lumley and Liu, 2006).

3.4. Personal exposure modeling

Table 4 shows the central-site infiltration factor (A, range: -0.07–2.05) that represents a combined effect of the outdoor spatial variation and infiltration estimated from the RCS model for each home, with the negative A having R^2 among the poorest model fits (<0.03). Thirteen of the 28 residences had low R^2 values (<0.2), suggesting that the central site $PM_{2.5}$ was a poor predictor for home outdoor $PM_{2.5}$ for these homes. Conventionally, the ratio of personal to home outdoor sulfur is used as the "true" ambient contribution fraction of $PM_{2.5}$ (α_{sulfur}), assuming that sulfur is a tracer of outdoor $PM_{2.5}$ (Sarnat et al., 2002; Allen et al., 2003; Ebelt et al., 2005). We evaluated our estimated ambient contribution fraction from the RCS model (α_{RCS}, Eq. (2)) against the α_{sulfur} calculated with the personal and central-site sulfur measurements. This was also an indirect verification of our A estimates. Due to the observed spatial variation of outdoor $PM_{2.5}$, our α_{sulfur} and α_{RCS} are not bounded by 1. The correlation between α_{sulfur} and α_{RCS} ($N = 18$) was 0.40, which increased to 0.87 when limited to residences with an R^2 for the RCS model greater than 0.2 ($N = 8$, A ranged from 0.21 to 2.05).

The estimated 12-h exposures to ambient-generated (E_a) and agricultural burning PM (E_b) based on the A estimates are shown in Table 5. Due to the incomplete TAD information, three subjects (i.e. B06, B15 and B23) had relatively small sample size. The mean E_a ranged from 3.7 to 20.8 $\mu g\, m^{-3}$ and the mean E_b ranged from 1.2 to 6.7 $\mu g\, m^{-3}$. The E_b was on average 33% of the E_a (SD = 14%, range = 2–68%). The GLM results show that after controlling for the subject effect ($p<0.01$), the E_b was on average 1.4 $\mu g\, m^{-3}$ higher during the episodes than non-episodes ($p<0.01$). The significant subject effect for E_b suggested that the central-site measurements would not capture individual variations in exposure.

The E_b estimated based on the RCS model, time–activities, and source apportionment results averaging over 24 h correlated with the 24-h average

Table 4
The particle central-site infiltration factor (A) estimated from the random component superposition model and particle infiltration efficiency (F_{inf}) estimated from the recursive model (sorted by RCS model's R^2 values)

ID	RCS model			RM
	A	R^2	N	F_{inf}
B15	0.71	0.66	54	[a]
B02	1.27	0.66	54	0.79
B30	0.61	0.66	56	0.78
B14	0.81	0.63	20	[a]
B21	1.11	0.59	54	0.63
B24	0.85	0.58	56	0.94
B01	1.25	0.57	61	0.73
B10	0.77	0.50	41	0.90
B06	0.48	0.46	46	0.49
B16	0.50	0.45	54	0.58
B03	0.48	0.41	50	[a]
B23	0.44	0.41	26	0.25
B19	0.49	0.39	46	0.70
B09	2.05	0.36	29	[a]
B13	0.21	0.30	54	0.71
B29	0.33	0.16	48	0.57
B20	0.29	0.15	54	0.55
B17	0.28	0.11	16	0.45
B22	0.17	0.11	53	0.56
B04	0.19	0.07	54	0.66
B07	0.29	0.04	51	0.69
B08	0.08	0.04	41	0.62
B26	-0.07	0.03	50	0.61
B27	0.28	0.02	30	[a]
B28	0.03	0.01	14	0.49
B31	0.07	0.01	54	0.51
B18	-0.03	0.00	33	0.41
B33	0.02	0.00	44	0.54

[a]No qualified data for the recursive model (see Appendix A).

Table 5
Summary of the estimated personal exposures to $PM_{2.5}$ from outdoor sources (E_a) and from agriculture burning smoke (E_b)

ID	N	E_a ($\mu g\,m^{-3}$)				E_b ($\mu g\,m^{-3}$)			
		RCS		RM		RCS		RM	
		Mean	(SD)	Mean	(SD)	Mean	(SD)	Mean	(SD)
(a)									
B01	71	14.0	(7.1)	8.7	(3.9)	4.6	(3.2)	2.8	(1.7)
B02	78	14.1	(7.1)	10.9	(3.9)	4.7	(3.2)	3.6	(2.0)
B03	80	10.7	(4.8)	9.8	(4.6)	3.5	(2.2)	3.2	(2.1)
B06	18	7.8	(3.6)	7.4	(3.3)	2.1	(1.1)	2.0	(1.1)
B09	77	20.8	(12.5)	9.5	(4.9)	6.7	(5.4)	3.0	(2.2)
B10	82	9.3	(4.5)	8.5	(4.4)	3.0	(2.0)	2.7	(2.0)
B13	89	7.4	(4.3)	9.0	(4.6)	2.4	(1.6)	2.9	(2.0)
B14	59	9.3	(4.4)	8.7	(4.4)	2.8	(1.6)	2.6	(1.6)
B15	17	10.4	(4.9)	10.6	(5.1)	2.8	(2.0)	2.8	(2.0)
B16	90	7.7	(3.7)	7.5	(3.3)	2.5	(1.6)	2.4	(1.3)
B19	66	7.9	(3.8)	10.2	(3.9)	2.8	(1.9)	3.6	(2.2)
B21	70	12.7	(6.0)	9.4	(3.9)	4.3	(2.8)	3.2	(2.2)
B23	14	6.5	(2.3)	4.2	(2.6)	2.2	(0.6)	1.3	(0.7)
B24	87	7.4	(3.5)	9.2	(4.6)	2.4	(1.4)	3.0	(2.1)
B30	44	9.4	(3.6)	11.7	(3.6)	3.4	(1.7)	4.3	(2.1)
(b)									
B04	89	5.4	(4.6)	9.8	(4.0)	1.7	(1.5)	3.1	(1.8)
B07	87	6.4	(4.0)	9.9	(3.9)	2.0	(1.4)	3.1	(1.9)
B08	89	4.4	(4.8)	7.3	(4.4)	1.4	(1.6)	2.3	(1.8)
B17	82	6.5	(4.0)	8.1	(3.6)	2.1	(1.9)	2.7	(1.9)
B18	78	4.9	(5.0)	8.3	(3.5)	1.6	(1.7)	2.8	(1.5)
B20	66	5.0	(2.5)	5.9	(3.0)	1.6	(1.0)	1.8	(1.2)
B22	42	5.1	(3.0)	9.4	(2.9)	1.7	(1.0)	3.4	(1.5)
B26	76	5.6	(6.2)	10.1	(4.1)	2.0	(2.7)	3.4	(2.2)
B27	79	7.3	(4.7)	10.7	(5.2)	2.5	(2.1)	3.6	(2.4)
B28	71	3.7	(3.6)	8.3	(3.0)	1.2	(1.2)	2.8	(1.4)
B29	62	6.9	(4.3)	7.5	(4.4)	2.3	(1.7)	2.6	(1.9)
B31	67	5.2	(3.8)	7.3	(3.6)	1.7	(1.3)	2.5	(1.6)
B33	44	3.8	(3.1)	7.3	(2.9)	1.3	(1.1)	2.6	(1.4)

(a) RCS's $R^2 > 0.2$; (b) RCS's $R^2 < 0.2$.

personal LG measurements ($r = 0.51$). The correlation increased to 0.58 after excluding homes with an R^2 for RCS smaller than 0.2 (denoted as open circle in Fig. 3a). This moderate correlation is due to the assumption of a uniform agricultural burning contribution across subjects. Eq. (3) assumed that the ratio of personal E_b to E_a is identical to the proportion of vegetative burning PM in total outdoor $PM_{2.5}$ at the central site, regardless of home locations and the contribution from residential wood burning. This proportion was fixed for any given 12-h period although the actual proportion might vary by location or subject. In reality, valley residents may receive different agricultural burning exposure from those residing on hill. The uncertainties in the CMB estimates also partially accounted for this moderate correlation. However, as it was expensive to measure daily personal exposure to LG on every subject, the estimated E_b served as an informative estimate of exposure to PM originated from agricultural burning smoke. Personal LG of the same 24 samples in Fig. 3a (filled circle) had a higher correlation with the central-site LG ($r = 0.84$, Fig. 3b). This relatively high correlation does not necessary imply an unbiased exposure estimate for individual subjects as personal LG exposure scattered substantially around the central-site measurements (Fig. 3b). Thus, the central-site LG might provide a good index for average population exposure in ecological studies but may not be representative of individual subjects' exposure to LG in a panel study.

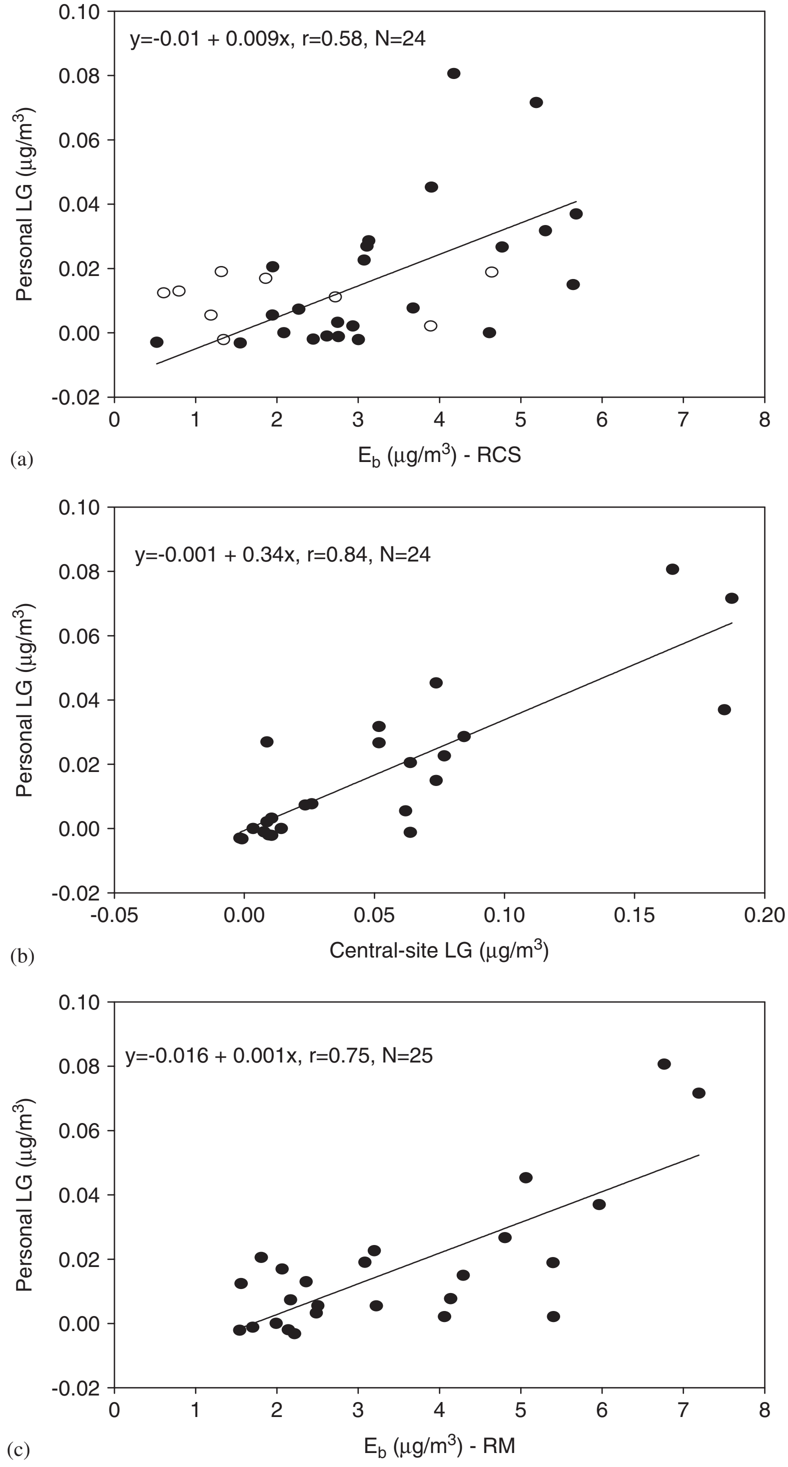

Fig. 3. (a) Comparison between personal LG and E_b estimated from the RCS model. Filled circle: residences with RCS's $R^2 > 0.2$. Open circle: residences with RCS's $R^2 < 0.2$. The regression line is fitted for the filled circle. (b) Comparison between personal and central-site LG. (c) Comparison between personal LG and E_b estimated from the RM.

3.5. Sensitivity analysis

The correlation between our E_b estimates and the LG measurements was reasonable, even with the presumption of $A = 1$ for the school and indoor microenvironments other than the home. We conducted a sensitivity analysis by changing the A values to 0.71 and 0.48, which were the 50th and 25th percentile, respectively, of the A estimates at residences with an R^2 for RCS larger than 0.2. The correlation between E_b and the LG measurements did not change much ($r = 0.55$ and 0.52), supporting our previous approximation of $A = 1$ at these two microenvironments.

We also tested the E_a and E_b estimates from the RCS model against those estimated from the RM, which was intended for estimating individual F_{inf}. Table 4 shows the F_{inf} estimated from the RM (Eq. (4)). Although A incorporated the spatial variation effect while F_{inf} did not, they correlated reasonably well ($r = 0.39$, $N = 11$) when limited to residences with an R^2 for RCS higher than 0.2. The ambient contribution fraction estimated from the RM (α_{RM}) could not predict α_{sulfur} ($r = -0.21$, $N = 15$) due to two factors. First, α_{RM} is bounded by one while α_{sulfur} is not. Thus, these two values may represent different levels of spatial information. Second, there was no indicator (such as the R^2 for the RCS model) for the goodness of the model fit for the RM to screen the non-representative F_{inf} estimates. The R^2 obtained during the linear regression process in Eq. (4) is not a good indicator because of the autocorrelation of the time series data in the RM.

Surprisingly, the E_b estimates from the RM correlated well with the personal LG measurements ($r = 0.75$, $N = 25$, Fig. 3c). While the RCS generally performed better than the RM in terms of the α estimates, the RM outperformed the RCS model when it comes to estimating E_b. This discord can be attributed to the fact that the $\alpha_{model}/\alpha_{sulfur}$ comparisons and E_b/LG comparisons were performed on different subject-days since few samples were analyzed for both sulfur and LG. Matched samples without missing α and E_b values were too limited ($N = 8$) for comparisons. Furthermore, $PM_{2.5}$ from all outdoor sources (with sulfur as the tracer) and $PM_{2.5}$ originated from biomass burning (with LG as the tracer) may have different spatial distribution, as well as different chemical and physical characteristics. The difference on spatial distribution is evident in the observed differences in the range of α_{LG} (0.46 ± 0.79) as compared with α_{sulfur} (0.75 ± 0.32). The correlation between α_{LG} and α_{sulfur} was 0.79, which dropped to 0.23 after removing one influential point of $\alpha_{LG} = 3.1$ (Fig. 4). Both regression slopes were significantly different from 1. Long and Sarnat (2004) analyzed infiltration efficiencies for various

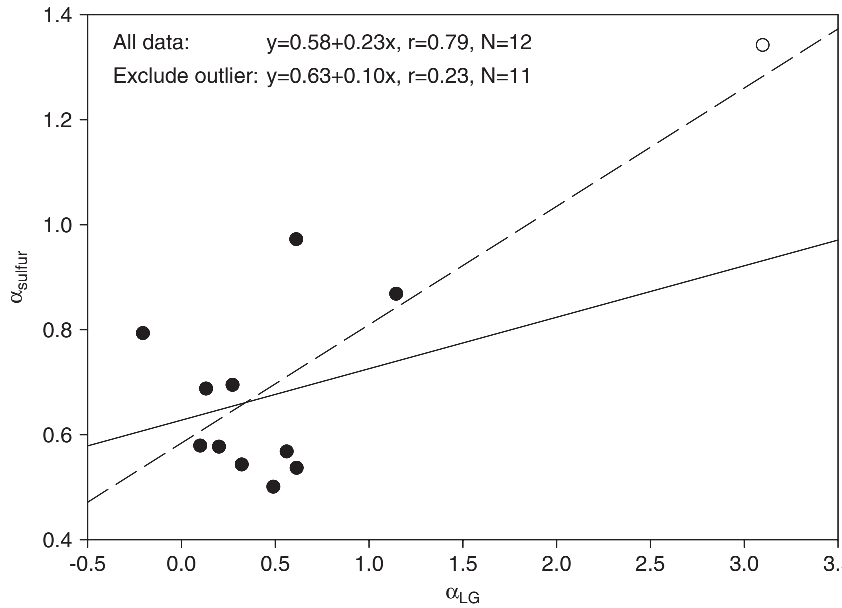

Fig. 4. Comparison between the ambient contribution fraction (α) estimated from the sulfur tracer and LG. Each point represents one subject-day observation (from 4 subjects).

elemental constituents in $PM_{2.5}$ and reported various infiltration behaviors among the elements. Thus, the A or F_{inf} estimated using the total $PM_{2.5}$ measurements may not represent the true A or F_{inf} for the $PM_{2.5}$ constituents originated from biomass burning smoke. To improve the E_b estimates, future studies should consider obtaining a better estimate of the infiltration efficiency for biomass related $PM_{2.5}$ by collecting both indoor and outdoor LG samples at subjects' homes. The National Research Council also suggested future studies focus on characterizing contribution fraction for specific PM components (National Research Council (NRC), 2004).

4. Conclusions

We found that during the agricultural burning season in Pullman, outdoor $PM_{2.5}$ exhibited a significant spatial variation, with the highest $PM_{2.5}$ concentrations occurring in the valley areas. The best predictors for the spatial variation included central site PM, elevation, and outdoor temperature. The mean personal $PM_{2.5}$ exposure was $13.8\,\mu g\,m^{-3}$ and was higher during the episodes than non-episodes. The personal LG and OC exposures were higher during episodes than during non-episodes, although significant between-subject variations were also observed. The cross-sectional correlation between the central-site and personal $PM_{2.5}$ concentrations (excluding outliers) in our study was higher than those reported previously, probably due to the close proximity of subjects' workplaces and residences to the central site. However, since it was found that subjects had different activity patterns during episodes, one should be cautious when applying central-site measurements as estimates of individual exposures in a cohort health study.

The mean personal exposure to $PM_{2.5}$ originated from agricultural burning smoke ranged between 1.2 and $6.7\,\mu g\,m^{-3}$ and between 1.3 and $4.3\,\mu g\,m^{-3}$ according to the RCS and RM, respectively. The E_b estimates also correlated with the personal LG measurements ($r = 0.51$ and 0.75 for the RCS and RM, respectively). These moderate correlations were probably due to the differences in infiltration between total $PM_{2.5}$ and vegetative burning related $PM_{2.5}$, the spatial variation of the agricultural burn smoke, the uncertainties in the CMB modeling estimates, and the limited range of E_b in this study. The small sample size of personal LG measurements prevents a comparative and conclusive assessment of the model performance. Although a high correlation ($r = 0.84$) between the personal and central-site LG was found, it did not imply an unbiased exposure estimate for individuals. Thus, the central-site measurements may better serve as the exposure index in an ecological study than in a panel study. We recommend future studies collect more microenvironmental samples to model the E_b and more personal samples to validate the E_b estimates.

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Appendix A. Data selection for the RM

The RM was performed from the 10 best consecutive days of data collected during the 30 monitoring days at each residence, based on the following criteria (in order of priority): (1) Nephelometer was used and its nighttime median hourly ratio of indoor to outdoor $PM_{2.5} < 1$ ($N = 8$ residences); (2) pDR was used and its nighttime median hourly ratio of indoor to outdoor $PM_{2.5} < 1$ ($N = 9$); (3) median hourly ratio of indoor pDR measurements to outdoor $PM_{2.5}$ during the best 10 consecutive days < 1 ($N = 2$); and (4) median nighttime hourly ratio of indoor pDR measurements to central-site $PM_{2.5}$ during the best 10 consecutive days < 1 ($N = 4$). Only the best 10 consecutive days of data, rather than all available data, are used for RM modeling because of the uncertainties in the light scattering efficiency in the pDR and because our spatial model may not be accurate at 1-h resolution. In addition, occasional local outdoor sources would not be reflected in the central site measurements.

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