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Impact of microenvironments and personal activities on personal PM2.5 exposures among asthmatic children

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Complete List of Authors:	Van Ryswyk, Keith; Health Canada, Air Health Science Division Wheeler, Amanda; Health Canada, Air Health Science Division Wallace, Lance; Consultant Kearney, Jill; Health Canada, Air Health Science Division You, Hongyu; Health Canada, Air Health Science Division Kulka, Ryan; Health Canada, Air Health Science Division Xu, Iris Xiaohong; University of Windsor, Department of Civil and Environmental Engineering
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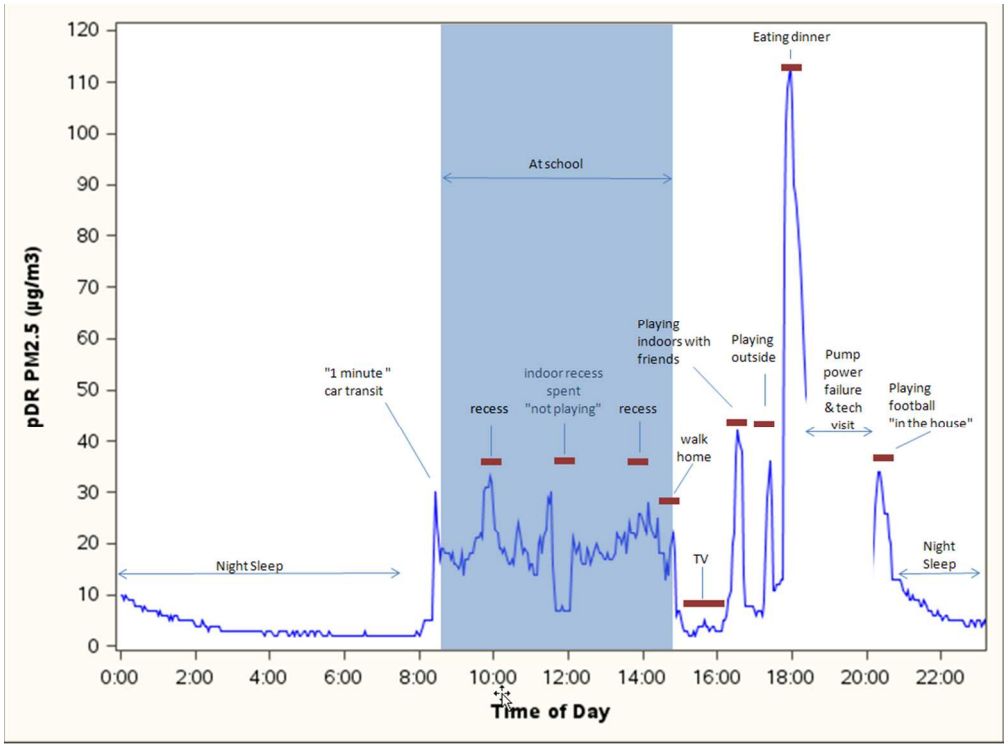

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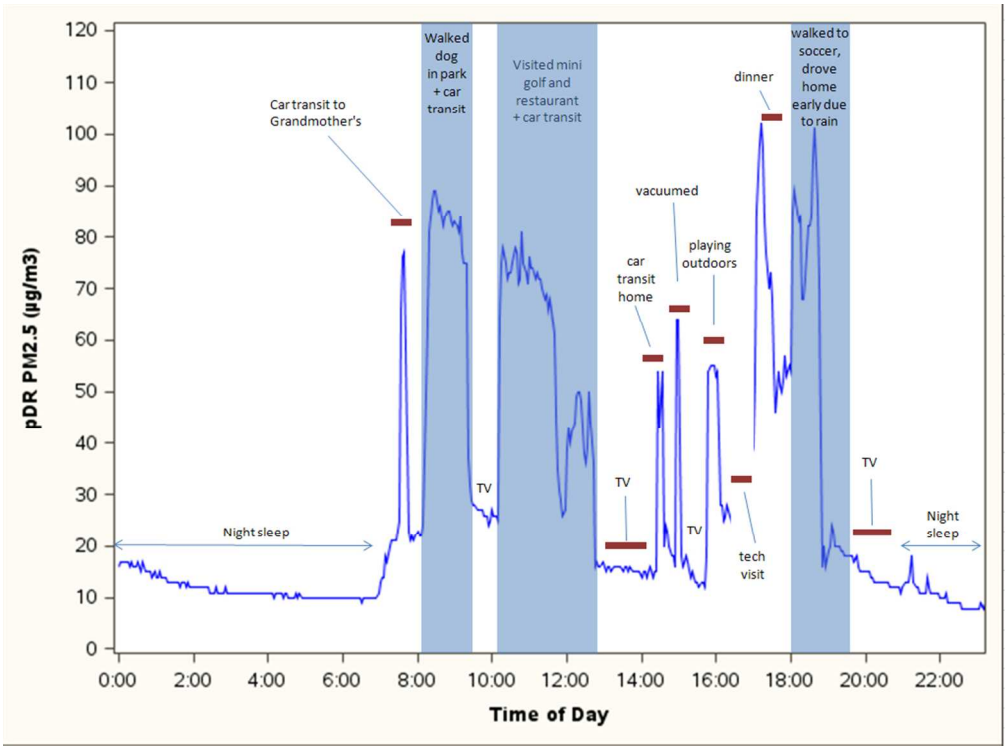

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Table 1: Number of study participants with complete data

Study Population		Winter	Summer
Age			
	10	17	15
	11	16	13
	12	8	6
	13	0	1
Ethnicity			
	Caucasian	38	33
	Other	3	2
Gender			
	Female	16	13
	Male	25	22
Heating Type			
	Electric	1	1
	Forced air	39	32
	Hot water	1	2
Home Type			
	Detached	35	32
	Rowhouse	4	2
	Duplex/triplex	2	1
Cooking Fuel			
	Electric	29	26
	Gas	12	9

125x190mm (96 x 96 DPI)

Table 2: Mean daily percent of time and exposure associated with excursions in $PM_{2.5}$.

Season	N 3-min data	$PM_{2.5}$ GMDA µg/m³	$PM_{2.5}$ MDA[1] µg/m³	excursion = $PM_{2.5}$ > MDA+1SD		excursion = $PM_{2.5}$ > MDA+2SD		excursion = $PM_{2.5}$ > MDA+3SD	
				% time	% exposure	% time	% exposure	% time	% exposure
Winter	68811	9.5(1.5)	11.2 (4.9)	20.8 (6.2)	58.9 (10.4)	14.5(6.2)	48.5(13.1)	10.5(5.5)	40.6(13.3)
Summer	59582	10.3(1.8)	13.7 (8.1)	18.0 (6.7)	54.6 (10.4)	12.5(5.5)	45.1(13.1)	8.9(5.3)	37.5(15.7)

standard deviation in parentheses

[1] Mean Daily Average (arithmetic)

287x72mm (96 x 96 DPI)

Table 3: Microenvironmental percent time, $PM_{2.5}$ exposure and percent contribution to total exposure

Season	Microenvironment	Participants occupying microenvironment	n 30 minute periods	$PM_{2.5}$ AM (SD) ($\mu g/m^3$)	$PM_{2.5}$ GM (GSD) ($\mu g/m^3$)	Mean % Time (SD)	Mean % Daily Exposure (SD)[1]
Winter n=41	Indoors at Home	41	4688	8.9 (5.9)	6.4 (1.8)	67.1 (12.7)	51.7 (14.8)
	Outdoors at Home	16	49	16.6 (11.7)	12.3 (1.9)	0.6 (1.1)	6.1 (5.2)
	In transit	39	325	17.2 (8.6)	13 (1.5)	4.4 (2.3)	9.0 (4.8)
	AtWork/School	40	1304	16.9 (7.0)	14.4 (1.5)	17.7 (5.9)	38.6 (11.7)
	Outdoors away from Home	36	173	16.5 (9.7)	12.3 (2.0)	2.4 (2.0)	6.6 (4.4)
	Indoors away from Home	36	361	17.1 (16.8)	12.3 (2.0)	5.3 (5.2)	17.2 (10.6)
Summer n=35	Indoors at Home	35	4576	11.5 (7.7)	8.3 (1.9)	72.3 (22.6)	66.3 (19.0)
	Outdoors at Home	29	284	18.3 (12.4)	11.9 (2.1)	4.3 (4.9)	12.1 (9.1)
	In transit	31	230	19.3 (12.6)	13 (2.2)	3.5 (3.0)	8.9 (5.5)
	AtWork/School	1	20	39.6 (.)	31.9 (.)	0.3 (1.8)	32.5 (.)
	Outdoors away from Home	21	243	23.2 (23.7)	14.4 (2.4)	3.9 (5.0)	18.5 (11.5)
	Indoors away from Home	32	620	25.6 (41.5)	12.5 (2.4)	10.3 (11.8)	23.4 (18.3)

[1] represents data of microenvironment occupying participants only.

292x129mm (96 x 96 DPI)

Table 5: Summary statisitics of daily percent time, personal $PM_{2.5}$ exposures and percent contributions of reported personal activities

Activity	Winter (n=41)						Summer (n=34)					
	N IDs	N 30min periods	AM(SD) $PM_{2.5}$[1] ($\mu g/m^3$)	GM(GSD) $PM_{2.5}$[1] ($\mu g/m^3$)	% Time (SD)	% Daily Exposure (SD)[2]	N IDs	N 30min periods	AM(SD) $PM_{2.5}$[1] ($\mu g/m^3$)	GM(GSD) $PM_{2.5}$[1] ($\mu g/m^3$)	% Time (SD)	% Daily Exposure (SD)[2]
Night sleep	41	3109	5.4 (3.6)	3.8 (2.0)	44.5 (8.2)	21.8 (9.1)	35	2788	8.6 (6.4)	5.2 (2.4)	44.7 (11.7)	30.5 (10.9)
Attending F/T school	40	1156	16.9 (6.9)	14.4 (1.5)	16.0 (5.7)	34.7 (10.6)	0	0			0 (0)%	
Homework	27	126	15.2 (19.1)	9.2 (2.2)	1.8 (2.7)	6.0 (4.4)	0	0			0 (0)%	
Watching TV	40	455	14.1 (15.8)	8.2 (2.4)	6.4 (4.8)	11.8 (9.8)	35	615	20.2 (32.2)	10.4 (2.3)	9.3 (8.1)	13.4 (9.7)
Eating	40	308	16.9 (10.8)	11.8 (1.8)	4.3 (2.1)	8.1 (5.4)	32	244	19.8 (17.0)	10.5 (2.6)	3.9 (2.3)	7.0 (3.2)
Computer use	19	104	11.1 (10.4)	7.7 (2.1)	1.6 (2.6)	6.9 (5.4)	22	275	12.3 (8.3)	8.2 (2.3)	4.6 (6.5)	10.7 (8.2)
Thinking/relaxing	27	159	8.8 (5.5)	4.2 (7.1)	2.7 (4.6)	5.6 (5.0)	25	210	12.0 (8.5)	7.6 (2.2)	3.0 (4.6)	5.6 (5.2)
Outdoor playing	28	132	17.2 (7.6)	13.6 (1.7)	1.8 (1.8)	7.3 (3.7)	25	233	18.2 (15.1)	11.3 (2.5)	3.5 (4.4)	12.2 (10.2)
Active sports	20	131	16.8 (12.5)	10.9 (2.7)	1.7 (2.1)	13.4 (11.0)	22	220	23.0 (18.1)	15.2 (2.1)	3.4 (4.3)	16.6 (9.7)
Car transit	31	130	14.8 (11.9)	11.5 (1.8)	1.8 (1.9)	6.0 (3.7)	27	178	19.6 (13.6)	12.9 (2.4)	2.6 (2.6)	8.6 (4.8)
Games	19	96	16.6 (14.2)	11.4 (2.0)	1.3 (2.1)	14.2 (17.6)	20	193	17.7 (7.7)	9.0 (2.3)	3.0 (4.7)	9.2 (7.6)
Indoor playing	14	97	25.2 (24.7)	18.1 (2.1)	1.5 (3.4)	13.0 (9.6)	21	136	23.3 (46.7)	10.3 (2.6)	2.1 (3.0)	10.2 (10.3)
Visiting	17	99	18.1 (26.9)	10.9 (2.5)	1.2 (2.0)	15.8 (16.3)	16	115	22.9 (32.6)	5.9 (12.5)	1.9 (4.1)	15.1 (15.8)
Personal hygiene	34	142	11.4 (9.8)	6.9 (2.8)	1.9 (1.6)	3.3 (1.7)	20	67	32.2 (71.2)	10.7 (2.5)	1.0 (1.3)	4.3 (3.1)
Movies/videos	11	51	9.3 (9.9)	5.4 (3.3)	0.7 (1.3)	7.9 (5.0)	18	134	14.1 (7.0)	11.7 (1.6)	2.2 (2.9)	11.1 (6.9)
Walking	22	93	16.2 (9.7)	11.1 (2.1)	1.3 (1.6)	5.6 (3.7)	8	15	22.3 (20.8)	15.4 (2.3)	0.2 (0.6)	3.9 (3.2)
Read books	9	33	9.3 (3.0)	8.1 (1.4)	0.4 (1.0)	3.0 (2.2)	10	62	11.0 (8.1)	6.9 (2.7)	1.1 (2.8)	5.9 (5.4)
Shopping for clothes/household items	11	24	19.6 (28.8)	11.7 (2.5)	0.4 (0.8)	10.9 (15.6)	20	67	20.8 (15.6)	14.1 (2.6)	1.1 (1.8)	9.4 (11.6)
Biking	1	5	25.2 (.)	23.7 (.)	0.1 (0.5)	5.7 (.)	13	50	19.2 (14.4)	14.0 (2.4)	0.9 (2.0)	7.9 (4.8)
(Other n = 51)	40	450	19.3 (20.7)	11.8 (2.0)	6.2 (3.9)	11.8 (7.4)	34	372	21.7 (23.0)	11.5 (2.6)	5.8 (3.6)	11.9 (6.3)

[1] As reported by the pDR.

[2] represents data of activity performing participants only.

307x148mm (96 x 96 DPI)

Table 4: Differences in personal exposure measured by the pDR in microenvironments compared with the referrent microenvironment 'indoors at home' and determined by the Generalized Estimating Equation Models.

Season	Microenvironment	N participants	N 30-min periods	Model Estimate $PM_{2.5}$ (SE) ($\mu g/m^3$)	Concentration difference from the referent condition (95% CI)
Winter n = 41	Indoors at Home	41	4688	9.9 (0.8)	
	Outdoors at Home	16	49	10.9 (2.6)	0.9 (-3.0 - 4.9)
	In transit	39	325	14.5 (1.3)	4.6 (3.2 - 6.0)**
	At School	40	1304	14.5 (1.1)	4.6 (3.0 - 6.2)**
	Outdoors away from Home	36	173	14.5 (1.4)	4.6 (2.4 - 6.7)**
	Indoors away from Home	36	361	13.7 (1.3)	3.8 (1.7 - 6.0)**
Summer n = 35	Indoors at Home	35	4576	13.2 (1.3)	
	Outdoors at Home	29	284	16.4 (2.7)	3.2 (0.3 - 6.0)*
	In transit	31	230	15.5 (1.9)	2.2 (-0.3 - 4.8)
	At Work	1	20	25.9 (1.2)	12.6 (-1.0 - 26.2)
	Outdoors away from Home	21	243	23.5 (6.3)	10.3 (6.6 - 13.9)**
	Indoors away from Home	32	620	17.3 (1.9)	4.1 (1.3 - 6.9)*

Model p-values: p=0.016 (winter), p=0.041 (summer)

n 30 min periods = 6900 (winter) 5973 (summer)

**p<0.001, *p<0.05

266x175mm (96 x 96 DPI)

Table 6: Differences in personal exposure measured by the pDR during particle generating activities performed indoors at home determined by the Generalized Estimating Equation Models.

Season	Particle generating or resuspending personal activity	N participants	N 30-min periods	Model Estimate $PM_{2.5}$ (SE) ($\mu g/m^3$)	Concentration difference from the referent condition (95% CI)
Winter	Sedentary	41	954	10.7 (1.1)	
	Night sleep	41	3088	6.6 (0.7)	-4.1 (-5.7 - -2.4)**
	Food preparation	9	17	40.7 (15.4)	30.0 (23.5 - 36.5)**
	Cleaning house	8	22	20.3 (5.2)	9.6 (3.2 - 16.0)*
	Indoor playing	14	93	20.8 (3.1)	10.1 (6.3 - 13.8)**
	Eating	34	184	15.8 (2.3)	5.1 (3.0 - 7.1)**
	Personal hygiene	39	213	12.9 (2.7)	2.2 (0.2 - 4.1)*
	Dressing, etc.	19	32	13.9 (4.8)	3.2 (-1.2 - 7.6)
	Games	17	85	13.6 (2.5)	2.9 (-0.8 - 6.6)
Summer	Sedentary	35	1296	12.4 (1.5)	
	Night sleep	35	2613	10.3 (1.2)	-2.1 (-3.8 - -0.3)*
	Food preparation	7	19	19.9 (2.9)	7.5 (1.6 - 13.4)*
	Cleaning house	13	29	12.9 (3)	0.6 (-4.5 - 5.6)
	Indoor playing	20	111	24 (8.8)	11.6 (8.1 - 15.1)**
	Eating	32	212	15.4 (2.5)	3.0 (1.2 - 4.8)*
	Personal hygiene	26	101	17.9 (4.1)	5.5 (2.9 - 8.0)**
	Dressing, etc.	6	9	11.7 (2.4)	-0.7 (-8.5 - 7.1)
	Games	19	186	12.5 (1.4)	0.2 (-2.8 - 3.1)

Model R^2: p=0.001 (winter), p=0.032 (summer)

n 30 min periods = 4688 (winter)4576 (summer)

**p<0.001, *p<0.05

245x196mm (96 x 96 DPI)

Impact of microenvironments and personal activities on personal $PM_{2.5}$ exposures among asthmatic children

Keith Van Ryswyk, BSc[a], Amanda J. Wheeler, PhD[a], Lance Wallace, PhD [b], Jill Kearney, MSc [a], Hongyu You, MSc[a], Ryan Kulka, BEng[a], and Xiaohong Xu, PhD[c]

[a]Air Health Science Division, Health Canada, Ottawa, Ontario, Canada

[b]Consultant, Santa Rosa, California

[c]Dept. of Civil and Environmental Engineering, University of Windsor, Ontario, Canada

Corresponding Author:

Keith Van Ryswyk

269 Laurier Avenue West

AL4903C

Ottawa, K1A 0K9, Ontario, Canada

Tel: 613-952-4582

Fax: 613-948-8482

Email: Keith.VanRyswyk@hc-sc.gc.ca

ABSTRACT

Personal activity patterns have often been suggested as a source of unexplained variability when comparing personal particulate matter (PM) exposure to modeled data using central site or microenvironmental data. To characterize the effect of personal activity patterns on asthmatic children's personal $PM_{2.5}$ exposure, data from the Windsor, Ontario Exposure Assessment Study were analyzed. The children spent on average 67.1±12.7% (winter) and 72.3±22.6% (summer) of their time indoors at home where they received 51.7±14.8% and 66.3±19.0% of their $PM_{2.5}$ exposure, respectively. In winter, 17.7±5.9% of their time was spent at school where they received 38.6±11.7% of their $PM_{2.5}$ exposure. In summer, they spent 10.3±11.8% 'indoors away from home' which represented 23.4±18.3% of their $PM_{2.5}$ exposure. Personal activity codes adapted from those of the National Human Activity Pattern Survey and the Canadian Human Activity Pattern Survey were assigned to the children's activities. Of the over 100 available activity codes, 19 activities collectively encompassed nearly 95% of their time. Generalized estimating equation (GEE) models found that, while indoors at home, relative to daytime periods when sedentary activities were conducted, several personal activities were associated with significantly elevated personal $PM_{2.5}$ exposures. Indoor playing represented a mean increase in $PM_{2.5}$ of 10.1 $\mu g/m^3$ (95%CI 6.3-13.8) and 11.6 $\mu g/m^3$ (95%CI 8.1-15.1) in winter and summer, respectively, as estimated by the pDR.

Keywords: $PM_{2.5}$; pDR; Personal exposure; Childhood asthma; personal activity

INTRODUCTION

Several studies have found associations between fine particulate matter ($PM_{2.5}$) and pediatric asthma incidence (McConnell et al., 2010), symptom severity (Delfino et al., 1998; Slaughter et al., 2003), related hospital admissions (Li et al., 2011; Norris et al., 1999; Strickland et al., 2010), and decreased lung function (Gauderman et al., 2004; He et al., 2010; Kulkarni and Grigg, 2008; O'Connor et al., 2008). In contrast to adults, children represent an especially sensitive population to $PM_{2.5}$ exposure where the same personal exposure results in a higher uptake per unit body weight. Children also have a higher breathing rate at rest than adults and they have a more active lifestyle which further increases their exposure. The breathing rates of children aged under 12 years have been shown to increase by a factor of 2 and 4 during moderate and heavy physical activity, respectively (Marty et al., 2002). In the case of asthmatic children, their lower levels of antioxidant defenses in the endothelium layer of the lung (Kelly, 2003) further increase their susceptibility to air pollution. Lastly, it has been seen that the oxidative stress of air pollution impedes the process of pulmonary morphogenesis during childhood, resulting in decreased lung function that impacts quality of life in adulthood and old age (Gauderman et al., 2004).

Fixed-site monitor (FSM) data has been used extensively as a surrogate for personal exposure to ambient pollution in many air pollution health effect studies (Dales et al., 2008; Dockery, 2001; Gauderman et al., 2004; Li et al., 2011). In the case of $PM_{2.5}$, its regionally consistent nature allows for the assumption that the concentrations measured at a single FSM may be used as a relevant measure of ambient $PM_{2.5}$ for a large area of a community. Community-wide estimates of ambient $PM_{2.5}$ can also be estimated using dispersion and land use regression modeling methods. However, studies comparing personal exposure levels of $PM_{2.5}$ to that of FSMs often cite a wide variation of correlations across participants (Adgate et al., 2003; Brown et al., 2009; Crist et al., 2008; Wallace, 2000). In many of these comparisons, personal activity and exposure to indoor sources has been suggested as potential sources of the variability.

Personal PM exposures have also been compared to estimates of exposure from microenvironmental (ME) modeling (Liu et al., 2003; Ozkaynak et al., 1996; Wu et al., 2005; Yip et al., 2004). These studies demonstrated that microenvironment-specific exposure data can account for a good deal of personal PM variability in elderly populations where most of their time is spent at home and where sedentary lifestyles are common. Considerable unexplained variability remains with pediatric subjects alluding to the question of what effects personal activities and the different microenvironments of children have on their personal PM exposure.

The Windsor, Ontario Exposure Assessment Study (WOEAS) involved the monitoring of 48 asthmatic children for 5 consecutive days in both the winter and summer of 2006. Personal activities and times spent in various microenvironments were recorded in time activity diaries and personal $PM_{2.5}$ exposure was monitored using the pDR. This paper characterizes the effect of the children's personal activity patterns on their $PM_{2.5}$ exposure by analyzing differences in personal $PM_{2.5}$ exposure by microenvironment and personal activitiy. An understanding of the effect of personal activity on personal $PM_{2.5}$ exposure for children can help identify the major sources and inform policy measures designed to mitigate their exposure.

METHODS

The methods used in WOEAS are more fully documented elsewhere (Wallace et al., 2011; Wheeler et al., 2011a). Briefly, participants were selected from recruits of the Windsor Children's Respiratory Health study (Dales et al., 2008). Using information from that study, children between 10-13 years with doctor diagnosed asthma living in non-smoking residences were recruited for the WOEAS in both the winter and summer of 2006. In each season, 48 children were monitored for a period of five consecutive days, from Monday to Saturday; technicians visited the participants daily. Each season, six participants were monitored concurrently during each of the eight sampling weeks. Each five day sampling week began on a Monday evening at approximately 4PM and

ended the following Saturday afternoon. During each 5 day period, their personal $PM_{2.5}$ exposures were measured and information on their personal activity patterns collected.

Time Activity Diary Data

Participants were asked to report their personal activities and microenvironmental locations through the use of time activity diaries (TAD). For each 30 minute period in their five days of monitoring, participants recorded their activity in open text and indicated their location in one of six microenvironments. The six categories of microenvironment were indoors at home, outdoors at home, in transit, at work/school, outdoors away from home and indoors away from home. The category of 'at work/school' was designated as such on account of the two seasons of the study. In winter, this category was understood to represent time at school and in summer, time spent in employment (one child reported working in the summer). TAD data was entered into electronic form in duplicate and discrepancies resolved. Each personal activity was classified using codes adapted from those of the National Human Activity Pattern Survey (NHAPS) (Klepeis et al., 2001) and the Canadian Human Activity Pattern Survey (CHAPS) (Leech et al., 1996). Coding was performed in duplicate to ensure consistency in the categorization of the child's activity.

Personal $PM_{2.5}$ Exposure Data

The personal DataRAM (pDR) (ThermoScientific, Waltham, MA, USA) has been extensively used in the measurement of personal PM exposure (Quintana et al., 2001; Wallace et al., 2003; Wallace et al., 2006; Wu et al., 2005; Yip et al., 2004). The pDR, calibrated to a NIST particle standard, features a continuous and light weight method of measuring particle concentrations in the air. It uses a laser at 880nm to measure mass concentration. Each participant carried a pDR to continuously measure personal $PM_{2.5}$ over the five days using a three minute logging interval, to allow for the assignment of personal activities and microenvironments identified through the TADs to the recorded exposure data. The pDR was also equipped with a Harvard Personal Environmental

Monitor (PEM; Chempass, R+P / Thermo). This filterless PEM acted as a size selective inlet, restricting particulates greater than 2.5 microns in diameter from entering the optical chamber of the pDR. The pDR set up also included a battery operated pump set to 1.8 lpm as required for the PEM. A second PEM, operating at 4.0 lpm, was also included in the personal monitoring assembly. This was a daily $PM_{2.5}$ sample which provided a gravimetric measure to which the pDR data could be compared.

At the end of each 24 hour period, end flows were recorded and recalibrated if necessary. PDR data associated with end flows varying by +/- 20% from the 1.8 lpm target were invalidated. At the end of each daily sampling period, positive drift was measured by replacing the PEM inlet with a HEPA filter and recording the display value after 60 seconds. In the event of a positive drift greater than 1 $\mu g/m^3$, a record of it was made for correction during data processing and the pDR was re-zeroed. Negative drift was indicated by differences between the internal and external averages of each pDR log. Internally, the pDR will measure negative values in particulate concentration. These negative values were used in the integrated pDR average reported with each daily data log; however, negative data were recorded in the instrument output as zero. Any difference between the machine-recorded daily average and the daily average calculated from the continuous data values output by the instrument provided an indication of, and a correction factor for, negative drift.

The pumps and pDRs were carried by the participants in a backpack. Inlets were positioned on the shoulder strap to appropriately sample in the participants' breathing zone. Participants were instructed to keep the backpacks with them throughout their daily activities and to note in their diaries when this could not be done i.e. bathing or swimming.

The pDR data was averaged into 30 minute periods in order to match it with the TAD data. Each 30 minute period was required to have at least 70% of valid TAD and pDR data to be included for analysis. Also, each day was required to have at least 18 hours of combined pDR and TAD data. Finally, participants were required to contribute at least 2 days of data in a season.

Statistical Analysis

Data management and statistical analyses were carried out using SAS V. 9.2 within SAS EG V. 4.2. (SAS Institute, Cary, NC). Daily percent exposure for each category of personal activity and microenvironment was calculated with the following equation:

$$P_{kij} = \frac{C_{kij} \times F_{kij}}{\sum_{k=1}^{n} C_{kij} \times F_{kij}} \quad \text{(Equation 1)}$$

where P_{kij} represents the percent contribution of category *'k'* to the total exposure of participant *'i'* on day 'j', C_{kji} represents the average $PM_{2.5}$ concentration of category *'k'* for participant *'i'* on day 'j', and F_{kji} represents the fraction of time spent in category *'k'* for participant *'i'* on day 'j'. Arithmetic averages (across days) were calculated for percent time, $PM_{2.5}$ exposure and percent exposure by season, participant and category. These arithmetic means were used to calculate the arithmetic and geometric means by season and category (across participants).

GEE models were used to estimate differences in personal $PM_{2.5}$ in microenvironments and during personal activities while accounting for autocorrelation and clustering. Personal activity and microenvironment models were run separately. The GEE models can be represented by the following equation:

$$y = \beta_o + \sum \beta_i x_i + \varepsilon \quad \text{(Equation 2)}$$

where y represents the exposure, $ß_o$ represents the model intercept, which is the concentration of the referent condition in the model, $ß_i$ are the model coefficients, x_i represents the 0/1 indicator variables for the microenvironments or activities, and ε

represents the model error. These GEE model analyses were carried out using the SAS GENMOD procedure, with an identity link function and an AR(1) autoregressive correlation structure. Estimated concentrations for each category were obtained using the LSMEANS option.

The referent condition in each model represented one of the several categories of 'microenvironment' or 'personal activity'. As such, the referent condition is the category of 'microenvironment' or 'personal activity' to which all other categories are compared in the model output. Referent condition selection does not affect model results, however; it can affect the ease of result interpretation. In all models, the choice of the referent condition was made by considering the nature of the data. Assigning the category (of microenvironment or activity) with the lowest mean exposure level as the referent condition resulted in positive model parameter estimates for each category. When analyzing the effect of microenvironments on exposure, x_i represented microenvironments other than 'indoors at home', which was assigned as the referent condition. Personal activities associated with particle generation (ex.: cooking, personal hygiene) and particle resuspension (ex.: playing, cleaning house) were represented by x_i, in the activity model. Sedentary activities (ex. watching TV, playing computer)were assigned as the referent condition 'Sedentary'.

Several personal activity models were attempted. To control for the effect of microenvironments on exposure while estimating increases in personal $PM_{2.5}$ exposure by personal activity, activity models were developed using data exclusive to single microenvironments. GEE activity models for 'indoors at home' and outdoors were attempted. The outdoor model included data from the microenvironments 'outdoors at home' and 'outdoors away from home'. In the activity model representing data from 'indoors at home', the activity 'night sleep' was given its own category. To include this activity in the referent condition 'Sedentary' would introduce a diurnal effect as this

activity represents most night hours of each day when indoor exposures are typically lower.

RESULTS

A total of 51 children were included in the 2006 WOEAS sampling sessions. Forty-eight children diagnosed with asthma were recruited for the winter season with an additional three recruited for the summer season on account of drop out. A total of 41 winter and 35 summer participants produced the required amount of pDR and TAD data to be included in this analysis, 27 of which participated in both seasons. Most participants lived in detached homes with forced air ventilation, were predominantly Caucasian and between the ages of 10 and 13 (Table 1).

Quality Assurance

In total, 204 598 3-minute personal $PM_{2.5}$ measures from 51 participants over the two seasons were recorded in the study. Two sets of siblings were present in summer and one in the winter. Measures belonging to one sibling of each pair were removed to preserve the data's independence (n=8 704, 4.3%). Invalidation due to the end sample flow varying by over 20% resulted in the loss of 11 551(5.6%) measures. Data loss on account of pDR malfunction, including loss of battery power, pump failure and damaged pDRs exhibiting unnatural trends in data, accounted for 36 932(18%) measures of personal $PM_{2.5}$. Positive drift corrections were performed on approximately 6% of the data ranging from 2-6 $\mu g/m^3$. Negative drift corrections ranging from 2-4 $\mu g/m^3$ were applied to 0.3% of the records.

The data was converted into 12 997 30-minute means for combination with the TAD data. Minor remaining loss was accounted for by missing TAD data, the removal of all 30-minute periods during which field technicians were reported present by participants and the data sufficiency criteria of at least seven 3-min pDR measures per 30-min mean, 18 hours of data per sample day and two days for every five day sampling session. The resulting dataset included 12 873 30-minute periods from 41 and 35 participants in the

winter and summer sessions, respectively. Of these participants, 27 were sampled in both seasons. The number of days per participant ranged from two to five.

The performance of the pDR-1000 in this study is documented in Wallace et al, (2011). Due to its dependence on the index of refraction and the density of the aerosol mixture (Görner et al., 1995), the exposures measured by this unit are typically overestimated compared to gravimetric $PM_{2.5}$ measures. In comparison with the filter based $PM_{2.5}$ PEM data, the pDR over predicted $PM_{2.5}$ by 60% with an R^2 of 71.4% (Wallace et al., 2011). Each data point in this comparison represents 24 hours of personal $PM_{2.5}$ exposure, each representing a unique combination of time spent in various microenvironments. As different microenvironments have varying levels of relative humidity and particle properties, which can affect the relationship between these two methods of measuring $PM_{2.5}$, it is reassuring to see this consistent relationship without having adjusted for this factor. The data presented in this paper have not been adjusted to the gravimetric method as this bias does not affect the relative relationships of measurements in different microenvironments and personal activities. It should be noted that these levels are not to be interpreted as actual $PM_{2.5}$ exposure values.

3-min $PM_{2.5}$ Exposure

Examples of exposures for one participant during a calendar weekday during winter and summer are depicted in Figures 1 and 2, respectively. These time series plots represent the $PM_{2.5}$ exposures logged by the pDR in three minute intervals annotated with open text entries from the TADs. Figure 1 features a school day with the microenvironment 'at school' representing a baseline exposure of approximately 20 $\mu g/m^3$. This is notably higher than the other microenvironments encountered during the day. Also seen in this figure are short term peaks approximately 40 $\mu g/m^3$ while playing indoors and outdoors. The activity 'eating dinner' represents exposures above 100 $\mu g/m^3$. Figure 2 (a summer day for the same participant) also features highly variable patterns in exposure. Exposures of 75 $\mu g/m^3$ are seen while in transit and at a restaurant. In both figures,

baseline levels of $PM_{2.5}$ are seen to change by microenvironment and certain activities are associated with sharp 'peaks' or 'excursions' in $PM_{2.5}$ exposure. Table 2 presents the arithmetic and geometric average daily $PM_{2.5}$ exposures by season. It also presents percentages of time and exposure attributable to excursions in $PM_{2.5}$. Excursions are defined as levels that exceed the daily mean plus one, two and three standard deviations.

Microenvironmental times and $PM_{2.5}$ exposures

Descriptive statistics for daily values of personal $PM_{2.5}$ exposure as measured by the pDR, percent time, and percent exposure are presented in Table 3. Means of percent exposure received for each microenvironment are calculated only for those participants who spent time in those locations according to their TAD. The lowest exposures in both seasons occurred while participants were 'indoors at home'. As such, it was selected as the referent condition in the microenvironmental GEE models presented in Table 4. In winter, the children spent most of their time at home (67.1±12.7%) and at school (17.7±5.9%) where they received nearly all of their exposure (51.7±14.8% and 38.6±11.7%, respectively). One participant had no data representing time 'at school' as the two days of data were collected on a Saturday and a weekday statutory holiday.

GEE model estimates for winter revealed significant increases in $PM_{2.5}$ in all microenvironments except for 'outdoors at home' for which there was limited data. These increases were relatively equal in magnitude. In summer, the time no longer spent at school shifted to cause increases in times spent outdoors and indoors away from home. The substantial time spent 'indoors away from home' was noted as time spent in daycare, shopping and in restaurants. The outdoor environments represented a considerable percentage of exposure. Summer GEE model estimates of increases in $PM_{2.5}$ in microenvironments relative to 'indoors at home' revealed significant increases in all microenvironments apart from 'in transit' and 'at work'. The microenvironment 'at work' was represented by one participant who spent ten hours in employment.

'Outdoors away from home' was associated with a mean increase of 10.3 μg/m^3 (95%CI: 6.6-13.9) of $PM_{2.5}$.

Personal activity times and $PM_{2.5}$ exposures

Daily means of percent exposure represented by each activity were only calculated for participants who performed them. Of the over 100 activity codes adopted from the NHAPS (Klepeis et al., 2001) and CHAPS (Leech et al., 1996) surveys, 70 were used in coding the personal activities reported by the participants. Of these, nearly 95% of the children's time was represented by 19 of the codes in winter and 17 in summer. The means of $PM_{2.5}$ pDR exposure, percent time, and percent of daily exposures for these activities are presented in Table 5. Night sleep represented the majority of the children's time in both seasons. It was observed that the median sleeping hours were 9PM-7AM and 10PM-9AM in the winter and summer, respectively (not reported). Table 6 presents the winter and summer GEE models for estimating increases in personal $PM_{2.5}$ while engaging in particle generating/resuspending activities while indoors at home. Of the 70 identified personal activities in the dataset, 32 were conducted indoors at home. These activities were predominantly sedentary (night sleep, watching TV, homework, etc.). The referent condition 'Sedentary' represented all sedentary personal activities other than 'Night sleep'. The exposure levels between these two categories were significantly different in both seasons with night sleep representing the lower $PM_{2.5}$ exposures, revealing a degree of diurnal variation in both seasons. While the children were indoors at home, the highest average increases in personal exposure were seen when they were involved in food preparation and indoor playing. Food preparation (which also included cooking) elevated average exposures by 40.7 μg/m^3 (95%CI 23.5-36.5) and 7.5 μg/m^3 (95%CI 1.6-13.4) in winter and summer respectively. 'Indoor playing' was reported by more of the participants and increased personal exposure on average by 10.1 μg/m^3 (95%CI 6.3-13.8) in the winter and 11.6 μg/m^3 (95%CI 8.1-15.5) in the summer. 'Cleaning house' was associated with a significant average increase in exposure in winter only (9.6 μg/m^3 (95%CI 3.2-16.0)). Both in winter and summer,

'Eating' and 'personal hygiene' were reported by most participants and were associated with significant increases in their personal $PM_{2.5}$ exposure. 'Eating' was associated with average increases of 5.1 μg/m^3 (95%CI 3.0-7.1) and 3.0 μg/m^3 (95%CI 1.2-4.8) in winter and summer, respectively. This activity however does indicate a degree of reporting non-compliance by the participants as this activity was assuredly done by each of them every day. The GEE models representing data collected in both outdoor environments found no significant association between personal $PM_{2.5}$ exposure as measured by the pDR and personal activity. Therefore, no data on elevated levels of $PM_{2.5}$ by personal activity while outdoors are presented in this paper.

DISCUSSION

We found the microenvironmental profiles of the children in this study to be similar to time activity profiles reported in the U.S. National Human Activity Pattern Survey (NHAPS) and the Canadian Human Activity Pattern Survey (CHAPS) for respondents aged < 11 years (NHAPS n = 1,126; CHAPS n= 324) (Kleipis et al., 2001; Leech et al., 2002). For NHAPS and CHAPS, respectively, mean percent times spent 'indoors at home' (70.52±1.17, 72.33±2.38), 'outdoors' (4.2±0.5, 4.3±1.0), 'at school' (7.8±0.8, 5.7±1.2) and 'in vehicles' (3.6±0.3, 3.7±0.5) were comparable to our data when considering that their sampling methods included year round data collection and represented an oversampling of weekend days (Klepeis et al., 2001, Leech et al., 1996). With regards to the time spent in school, which was noted in the winter season only, the children spent an average of 67.1±12.7% of their time indoors at home and 17.7±5.9% of their time at school. These values are comparable to studies that have also monitored children during the school year (Noullett et al., 2006; Liu et al., 2003; Wu et al., 2005). Lastly, as the two seasonal sampling periods in this study were labelled as 'winter' and 'summer', they each also exclusively represented months where children were 'in school' and 'out of school', respectively. As the impact of 'in school' on average daily $PM_{2.5}$ exposure

was found to be substantial, it should be noted that it is a microenvironment occupied by children on a very consistent schedule ten months out of the year.

As can be seen in Table 3, the microenvironment 'indoors at home' had the lowest $PM_{2.5}$ concentration (ie. lowest exposure intensity); however, because participants spent most of their time there (67% winter, 72% summer), the greatest proportion of daily personal exposure occurred while indoors at home (52% winter, 66% summer). Comparatively, other microenvironments represented percentages of daily exposure much higher than those of time, revealing them to have much higher exposure intensities. Most notably, the outdoor microenvironments in both seasons had percentages of daily exposure 3 or 4 times higher than the percentages of time spent there. This situation may not be typical of most cities. In Wheeler et al., (2011b), the outdoor $PM_{2.5}$ levels were seen to exceed that of personal and indoor exposure and it was furthermore stated that this was not typical of most North American cities.

It's important to note that the microenvironments discussed in this paper can have different particle sources and with that represent different compositions and potential toxicities. Most notably, particulates of indoor and outdoor environments can represent different health risks. Outdoor environments represent particulates from combustion sources whereas indoor environments represent outdoor particulates that have infiltrated into the home and indoor sourced PM such as allergens and particulates resulting from personal activities. Estimates of the ambient fraction of indoor $PM_{2.5}$ exposure of the Windsor participants have been made using several infiltration methods (MacNeill et al., 2012). Indoor $PM_{2.5}$ exposure levels were 59% ambient in winter and 65% ambient in summer. Similarly, the increases in personal $PM_{2.5}$ associated with personal activities described in this paper can also represent particles of differing toxicities. The particles related to activities such as 'cleaning house' and 'indoor playing' represent particle characteristics conceivably different than those of 'personal hygiene' and 'food preparation', for example.

The impact of various microenvironments encountered in a day on personal PM exposure has been studied in comparisons of direct personal exposure measures to estimates using microenvironmental (ME) modeling (Liu et al., 2003; Ozkaynak et al., 1996; Wu et al., 2005; Yip et al., 2004). These models sum up time-weighted exposures from various microenvironments to predict personal PM exposure. Regressing the ME estimates against directly measured personal exposure quantifies how much variability in the personal exposure data can be accounted for by the microenvironmental measures. In Liu et al., (2003), this approach was used for 133 elderly and 33 asthmatic children in Seattle, WA. $PM_{2.5}$ exposure data for personal and other microenvironments were measured using integrated 24h gravimetric sampling. The ME model estimates explained 46% to 65% percent of the variability in the elderly personal $PM_{2.5}$ exposure data and 9% of the children's. It was suggested that the lack of predictive power in the case of the children was due to their active lifestyle, as it was noted that the elderly subjects were typically more sedentary and had lower variability in the microenvironments they occupied. The use of integrated sampling in each microenvironment likely weakened the model's power as the microenvironmental exposure data reflected time when the participants were not present. Wu et al. (2005) applied the ME model for 20 asthmatic children in Alpine, CA, USA. These methods were improved by the use of continuous monitoring in each microenvironment. This allowed the use of microenvironmental exposure data specific to the times in which the participants occupied them to be used in the model. Hourly ME estimates were calculated and averaged into daily $PM_{2.5}$ means. When regressed against the directly measured personal exposure data, 6% and 48% of the variability was explained for the data representing children-days spent 'in-school' and 'not-in-school', respectively. This improved approach yielded a decent degree of explained variability in the case of the 'not-in-school' group. The low R^2 of the 'in-school' group was suggested to be due to the use of exposure data collected in the 'indoors at home' microenvironment to represent time spent at school. The potential for difference in exposure between these

two microenvironments is seen in our own microenvironmental analysis where we found $PM_{2.5}$ exposures at school to be 4.6 μg/m^3 (95%CI 3.0-6.2) higher than indoors at home and contribute an average of 38.6±11.7% of daily $PM_{2.5}$ exposure.

The use of GEE models to estimate differences in personal PM exposure measured with the pDR during particle generating activities and between microenvironments has been attempted elsewhere (Allen et al., 2004; Quintana et al., 2001). In Allen et al, (2004) 38 monitoring events (the monitoring of a subject for a single 5 or 10 day session) included healthy, elderly adults (n=5), elderly patients with congestive heart failure (n=11), or with chronic obstructive pulmonary disease (n=10) and asthmatic children (n=2). A significant increase of 7.88 μg/m^3 in PM exposure was seen for the microenvironment 'at work' which was represented by one participant. The microenvironment 'in transit' was a moderately significant (p=0.07) 1.62 μg/m^3 average increase in personal exposure. Particle generating activities 'at school' were associated with average increases of 5.76 μg/m^3 (p<0.001) and 8.3 μg/m^3 (p<0.001) in personal $PM_{2.5}$ during class and recess, respectively. Cooking was associated with an increase of 5.46 μg/m^3 (p<0.05) in personal exposure. Quintana et al, (2001) measured the personal PM exposure of ten non-smoking adult volunteers for one week using the pDR. Using the GEE method, $PM_{2.5}$ concentrations in microenvironments designated as 'outdoors' were, on average, 37.3 μg/m^3 higher than 'indoors' (the referent condition in their model). In their personal activity GEE model, activities such as yard work, construction, and cooking were also seen to have significantly elevated exposures.

In this paper, the methods of determining the effect of microenvironments and personal activities on elevated particle exposures proved effective. These methods included: the measurement of trends in $PM_{2.5}$ exposure with the pDR, the collection of activity pattern data with TADs and the analysis of these combined data using the GEE method. However, some limitations are noted. The analysis was limited by an averaging time of 30 minutes imposed by the design of the TADs. This affects the analysis in several ways.

Changes in activity and microenvironments can easily vary within 30 minute time periods. In Figure 1, a car commute is described in open text as a 'one minute' commute. The result of this entry would be the assignment of the activity 'car transit' to a 30 minute time period thereby over estimating its duration. Furthermore, the 30 minute average of the three minute pDR data assigned to 'car transit' would represent three minutes of true private car exposure and 27 minutes of misclassified exposure. In this particular case where the exposures before and after the commuting event are lower, we have an underestimate of exposure for the microenvironment 'in transit'. Other examples of peaks lasting less than 30 minutes can be seen in Figures 1 and 2 implying that the elimination of this error with the use of higher temporal resolution TAD data may result in refined exposure estimates for activities and microenvironments. Aside from the issue of misclassification, a reduction in variability results by averaging the three minute pDR data into 30 minute periods. Daily maximum values for each participant-day were reduced by a mean factor of 2.0 (range 1.1-6.5) when converting the 3 minute data to 30 minute averaging periods. Mean coefficients of variance (CV) for each season and participant combination were calculated and reduced from 164% (range 43-840%) to 143% (range 43-658%) when averaged to 30 minute periods. Given a finer temporal resolution for our data, this variability could be preserved. This is difficult to implement especially for children as compliance in completing TAD is challenging.

To monitor time activity information at higher temporal resolutions, novel approaches have begun to emerge. Use of Global positioning system (GPS) tracking devices show promise for more accurate reporting of time-location patterns of children aged 3-5 than parent completed diaries (Elgethun et al., 2007). The Personal Digital Assistant (PDA) system for Activity Registration and Recording of Travel Scheduling (PARROTS) collects both GPS and personal activity data using default answers and predefined dropdown lists. This method features a reduction in participant burden, an increase in temporal resolution and has been found to decrease respondent error in comparison with paper diaries (Bellemans et al., 2008).

We combined time activity information collected with diaries and personal $PM_{2.5}$ exposure data collected with pDRs to relate personal exposure to microenvironments and activities. We found the effect of microenvironments and personal activities on elevated personal exposure to $PM_{2.5}$ to be significant. In winter, exposures while at school were found to be significantly elevated relative to indoors at home and constitute, on average, nearly 40% of the children's total daily exposure. We also found significant increases in personal $PM_{2.5}$ were attributed to personal activity while indoors at home. Although this panel study was conducted over a single year with a relatively small sample size of asthmatic children, who were not selected at random, their activity patterns are remarkably similar to those documented in national activity pattern surveys and other panel studies involving children. Understanding the sources of personal PM exposure and the significance of times spent near these sources is important when assigning exposure to populations and estimating the consequent health effects. Although centrally located fixed site monitoring data and microenvironmental models have some success with characterizing personal PM exposure, our findings indicate that the active lifestyle of children represents a significant factor in understanding their true $PM_{2.5}$ exposure.

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Impact of microenvironments and personal activities on personal $PM_{2.5}$ exposures among asthmatic children

Keith Van Ryswyk, BSc[a], Amanda J. Wheeler, PhD[a], Lance Wallace, PhD [b], Jill Kearney, MSc [a], Hongyu You, MSc[a], Ryan Kulka, BEng[a], and Xiaohong Xu, PhD[c]

[a]Air Health Science Division, Health Canada, Ottawa, Ontario, Canada

[b]Consultant, Santa Rosa, California

[c]Dept. of Civil and Environmental Engineering, University of Windsor, Ontario, Canada

Corresponding Author:

Keith Van Ryswyk

269 Laurier Avenue West

AL4903C

Ottawa, K1A 0K9, Ontario, Canada

Tel: 613-952-4582

Fax: 613-948-8482

Email: Keith.VanRyswyk@hc-sc.gc.ca

ABSTRACT

Personal activity patterns have often been suggested as a source of unexplained variability when comparing personal particulate matter (PM) exposure to modeled data using central site or microenvironmental data. To characterize the effect of personal activity patterns on asthmatic children's personal $PM_{2.5}$ exposure, data from the Windsor, Ontario Exposure Assessment Study were analyzed. The children spent on average 67.1±12.7% (winter) and 72.3±22.6% (summer) of their time indoors at home where they received 51.7±14.8% and 66.3±19.0% of their $PM_{2.5}$ exposure, respectively. In winter, 17.7±5.9% of their time was spent at school where they received 38.6±11.7% of their $PM_{2.5}$ exposure. In summer, they spent 10.3±11.8% 'indoors away from home' which represented 23.4±18.3% of their $PM_{2.5}$ exposure. Personal activity codes adapted from those of the National Human Activity Pattern Survey and the Canadian Human Activity Pattern Survey were assigned to the children's activities. Of the over 100 available activity codes, 19 activities collectively encompassed nearly 95% of their time. Generalized estimating equation (GEE) models found that, while indoors at home, relative to daytime periods when sedentary activities were conducted, several personal activities were associated with significantly elevated personal $PM_{2.5}$ exposures. Indoor playing represented a mean increase in $PM_{2.5}$ of 10.1 μg/m^3 (95%CI 6.3-13.8) and 11.6 μg/m^3 (95%CI 8.1-15.1) in winter and summer, respectively, as estimated by the pDR.

Keywords: $PM_{2.5}$; pDR; Personal exposure; Childhood asthma; personal activity

INTRODUCTION

Several studies have found associations between fine particulate matter ($PM_{2.5}$) and pediatric asthma incidence (McConnell et al., 2010), symptom severity (Delfino et al., 1998; Slaughter et al., 2003), related hospital admissions (Li et al., 2011; Norris et al., 1999; Strickland et al., 2010), and decreased lung function (Gauderman et al., 2004; He et al., 2010; Kulkarni and Grigg, 2008; O'Connor et al., 2008). In contrast to adults, children represent an especially sensitive population to $PM_{2.5}$ exposure where the same personal exposure results in a higher uptake per unit body weight. Children also have a higher breathing rate at rest than adults and they have a more active lifestyle which further increases their exposure. The breathing rates of children aged under 12 years have been shown to increase by a factor of 2 and 4 during moderate and heavy physical activity, respectively (Marty et al., 2002). In the case of asthmatic children, their lower levels of antioxidant defenses in the endothelium layer of the lung (Kelly, 2003) further increase their susceptibility to air pollution. Lastly, it has been seen that the oxidative stress of air pollution impedes the process of pulmonary morphogenesis during childhood, resulting in decreased lung function that impacts quality of life in adulthood and old age (Gauderman et al., 2004).

Fixed-site monitor (FSM) data has been used extensively as a surrogate for personal exposure to ambient pollution in many air pollution health effect studies (Dales et al., 2008; Dockery, 2001; Gauderman et al., 2004; Li et al., 2011). In the case of $PM_{2.5}$, its regionally consistent nature allows for the assumption that the concentrations measured at a single FSM may be used as a relevant measure of ambient $PM_{2.5}$ for a large area of a community. Community-wide estimates of ambient $PM_{2.5}$ can also be estimated using dispersion and land use regression modeling methods. However, studies comparing personal exposure levels of $PM_{2.5}$ to that of FSMs often cite a wide variation of correlations across participants (Adgate et al., 2003; Brown et al., 2009; Crist et al., 2008; Wallace, 2000). In many of these comparisons, personal activity and exposure to indoor sources has been suggested as potential sources of the variability.

Personal PM exposures have also been compared to estimates of exposure from microenvironmental (ME) modeling (Liu et al., 2003; Ozkaynak et al., 1996; Wu et al., 2005; Yip et al., 2004). These studies demonstrated that microenvironment-specific exposure data can account for a good deal of personal PM variability in elderly populations where most of their time is spent at home and where sedentary lifestyles are common. Considerable unexplained variability remains with pediatric subjects alluding to the question of what effects personal activities and the different microenvironments of children have on their personal PM exposure.

The Windsor, Ontario Exposure Assessment Study (WOEAS) involved the monitoring of 48 asthmatic children for 5 consecutive days in both the winter and summer of 2006. Personal activities and times spent in various microenvironments were recorded in time activity diaries and personal $PM_{2.5}$ exposure was monitored using the pDR. This paper characterizes the effect of the children's personal activity patterns on their $PM_{2.5}$ exposure by analyzing differences in personal $PM_{2.5}$ exposure by microenvironment and personal activitiy. An understanding of the effect of personal activity on personal $PM_{2.5}$ exposure for children can help identify the major sources and inform policy measures designed to mitigate their exposure.

METHODS

The methods used in WOEAS are more fully documented elsewhere (Wallace et al., 2011; Wheeler et al., 2011a). Briefly, participants were selected from recruits of the Windsor Children's Respiratory Health study (Dales et al., 2008). Using information from that study, children between 10-13 years with doctor diagnosed asthma living in non-smoking residences were recruited for the WOEAS in both the winter and summer of 2006. In each season, 48 children were monitored for a period of five consecutive days, from Monday to Saturday; technicians visited the participants daily. Each season, six participants were monitored concurrently during each of the eight sampling weeks. Each five day sampling week began on a Monday evening at approximately 4PM and

ended the following Saturday afternoon. During each 5 day period, their personal $PM_{2.5}$ exposures were measured and information on their personal activity patterns collected.

Time Activity Diary Data

Participants were asked to report their personal activities and microenvironmental locations through the use of time activity diaries (TAD). For each 30 minute period in their five days of monitoring, participants recorded their activity in open text and indicated their location in one of six microenvironments. The six categories of microenvironment were indoors at home, outdoors at home, in transit, at work/school, outdoors away from home and indoors away from home. The category of 'at work/school' was designated as such on account of the two seasons of the study. In winter, this category was understood to represent time at school and in summer, time spent in employment (one child reported working in the summer). TAD data was entered into electronic form in duplicate and discrepancies resolved. Each personal activity was classified using codes adapted from those of the National Human Activity Pattern Survey (NHAPS) (Klepeis et al., 2001) and the Canadian Human Activity Pattern Survey (CHAPS) (Leech et al., 1996). Coding was performed in duplicate to ensure consistency in the categorization of the child's activity.

Personal $PM_{2.5}$ Exposure Data

The personal DataRAM (pDR) (ThermoScientific, Waltham, MA, USA) has been extensively used in the measurement of personal PM exposure (Quintana et al., 2001; Wallace et al., 2003; Wallace et al., 2006; Wu et al., 2005; Yip et al., 2004). The pDR, calibrated to a NIST particle standard, features a continuous and light weight method of measuring particle concentrations in the air. It uses a laser at 880nm to measure mass concentration. Each participant carried a pDR to continuously measure personal $PM_{2.5}$ over the five days using a three minute logging interval, to allow for the assignment of personal activities and microenvironments identified through the TADs to the recorded exposure data. The pDR was also equipped with a Harvard ~~p~~Personal ~~e~~Environmental

~~m~~Monitor (PEM; Chempass, R+P / Thermo). This filterless PEM acted as a size selective inlet, restricting particulates greater than 2.5 microns in diameter from entering the optical chamber of the pDR. The pDR set up also included a battery operated pump set to 1.8 lpm as required for the PEM. A second PEM, operating at 4.0 lpm, was also included in the personal monitoring assembly. This was a daily $PM_{2.5}$ sample which provided a gravimetric measure to which the pDR data could be compared. ~~Concentrations were logged in three minute intervals.~~

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At the end of each 24 hour period, end flows were recorded and recalibrated if necessary. PDR data associated with end flows varying by +/- 20% from the 1.8 lpm target were invalidated. At the end of each daily sampling period, positive drift was measured by replacing the PEM inlet with a HEPA filter and recording the display value after 60 seconds. In the event of a positive drift greater than 1 $\mu g/m^3$, a record of it was made for correction during data processing and the pDR was re-zeroed. Negative drift was indicated by differences between the internal and external averages of each pDR log. Internally, the pDR will measure negative values in particulate concentration. These negative values were used in the integrated pDR average reported with each daily data log; however, negative data were recorded in the instrument output as zero. Any difference between the machine-recorded daily average and the daily average calculated from the continuous data values output by the instrument provided an indication of, and a correction factor for, negative drift.

The pumps and pDRs were carried by the participants in a backpack. Inlets were positioned on the shoulder strap to appropriately sample in the participants' breathing zone. Participants were instructed to keep the backpacks with them throughout their daily activities and to note in their diaries when this could not be done i.e. bathing or swimming.

The pDR data was averaged into 30 minute periods in order to match it with the TAD data. Each 30 minute period was required to have at least 70% of valid TAD and pDR data to be included for analysis. Also, each day was required to have at least 18 hours of combined pDR and TAD data. Finally, participants were required to contribute at least 2 days of data in a season.

Statistical Analysis

Data management and statistical analyses were carried out using SAS V. 9.2 within SAS EG V. 4.2. (SAS Institute, Cary, NC). Daily percent exposure for each category of personal activity and microenvironment was calculated with the following equation:

$$P_{kij} = \frac{C_{kij} \times F_{kij}}{\sum_{k=1}^{n} C_{kij} \times F_{kij}} \quad \text{(Equation 1)}$$

where P_{kij} represents the percent contribution of category *'k'* to the total exposure of participant *'i'* on day 'j', C_{kji} represents the average $PM_{2.5}$ concentration of category *'k'* for participant *'i'* on day 'j', and F_{kji} represents the fraction of time spent in category *'k'* for participant *'i'* on day 'j'. Arithmetic averages (across days) were calculated for percent time, $PM_{2.5}$ exposure and percent exposure by season, participant and category. These arithmetic means were used to calculate the arithmetic and geometric means by season and category (across participants).

GEE models were used to estimate differences in personal $PM_{2.5}$ in microenvironments and during personal activities while accounting for autocorrelation and clustering. Personal activity and microenvironment models were run separately. The GEE models can be represented by the following equation:

$$y = \beta_o + \sum \beta_i x_i + \varepsilon \quad \text{(Equation 2)}$$

where y represents the exposure, $ß_o$ represents the model intercept, which is the concentration of the referent condition in the model, $ß_i$ are the model coefficients, x_i represents the 0/1 indicator variables for the microenvironments or activities, and ε

represents the model error. These GEE model analyses were carried out using the SAS GENMOD procedure, with an identity link function and an AR(1) autoregressive correlation structure. Estimated concentrations for each category were obtained using the LSMEANS option.

The referent condition in each model represented one of the several categories of 'microenvironment' or 'personal activity'. As such, the referent condition is the category of 'microenvironment' or 'personal activity' to which all other categories are compared in the model output. Referent condition selection does not affect model results, however; it can affect the ease of result interpretation. In all models, the choice of the referent condition was made by considering the nature of the data. Assigning the category (of microenvironment or activity) with the lowest mean exposure level as the referent condition resulted in positive model parameter estimates for each category. When analyzing the effect of microenvironments on exposure, x_i represented microenvironments other than 'indoors at home', which was assigned as the referent condition. Personal activities associated with particle generation (ex.: cooking, personal hygiene) and particle resuspension (ex.: playing, cleaning house) were represented by x_i, in the activity model. Sedentary activities (ex. watching TV, playing computer)were assigned as the referent condition 'Sedentary'.

Several personal activity models were attempted. To control for the effect of microenvironments on exposure while estimating increases in personal $PM_{2.5}$ exposure by personal activity, activity models were developed using data exclusive to single microenvironments. GEE activity models for 'indoors at home' and outdoors were attempted. The outdoor model included data from the microenvironments 'outdoors at home' and 'outdoors away from home'. In the activity model representing data from 'indoors at home', the activity 'night sleep' was given its own category. To include this activity in the referent condition 'Sedentary' would introduce a diurnal effect as this

activity represents most night hours of each day when indoor exposures are typically lower.

RESULTS

A total of 51 children were included in the 2006 WOEAS sampling sessions. Forty-eight children diagnosed with asthma were recruited for the winter season with an additional three recruited for the summer season on account of drop out. A total of 41 winter and 35 summer participants produced the required amount of pDR and TAD data to be included in this analysis, 27 of which participated in both seasons. Most participants lived in detached homes with forced air ventilation, were predominantly Caucasian and between the ages of 10 and 13 (Table 1).

Quality Assurance

In total, 204 598 3-minute personal $PM_{2.5}$ measures from 51 participants over the two seasons were recorded in the study. Two sets of siblings were present in summer and one in the winter. Measures belonging to one sibling of each pair were removed to preserve the data's independence (n=8 704, 4.3%). Invalidation due to the end sample flow varying by over 20% resulted in the loss of 11 551(5.6%) measures. Data loss on account of pDR malfunction, including loss of battery power, pump failure and damaged pDRs exhibiting unnatural trends in data, accounted for 36 932(18%) measures of personal $PM_{2.5}$. Positive drift corrections were performed on approximately 6% of the data ranging from 2-6 $\mu g/m^3$. Negative drift corrections ranging from 2-4 $\mu g/m^3$ were applied to 0.3% of the records.

The data was converted into 12 997 30-minute means for combination with the TAD data. Minor remaining loss was accounted for by missing TAD data, the removal of all 30-minute periods during which field technicians were reported present by participants and the data sufficiency criteria of at least seven 3-min pDR measures per 30-min mean, 18 hours of data per sample day and two days for every five day sampling session. The resulting dataset included 12 873 30-minute periods from 41 and 35 participants in the

winter and summer sessions, respectively. Of these participants, 27 were sampled in both seasons. The number of days per participant ranged from two to five.

The performance of the pDR-1000 in this study is documented in Wallace et al, (2011). Due to its dependence on the index of refraction and the density of the aerosol mixture (Görner et al., 1995), the exposures measured by this unit are typically overestimated compared to gravimetric $PM_{2.5}$ measures. ~~Along with the pDR, each participant carried daily $PM_{2.5}$ personal environment monitors (PEM; Chempass System R&P/Thermo).~~ In comparison with the~~se~~ filter based $PM_{2.5}$ PEM data, the pDR over predicted $PM_{2.5}$ by 60% with an R^2 of 71.4% (Wallace et al., 2011). Each data point in this comparison represents 24 hours of personal $PM_{2.5}$ exposure, each representing a unique combination of time spent in various microenvironments. As different microenvironments have varying levels of relative humidity and particle properties, which can affect the relationship between these two methods of measuring $PM_{2.5}$, it is reassuring to see this consistent relationship without having adjusted for this factor. The data presented in this paper have not been adjusted to the gravimetric method as this bias does not affect the relative relationships of measurements in different microenvironments and personal activities. It should be noted that these levels are not to be interpreted as actual $PM_{2.5}$ exposure values.

3-min $PM_{2.5}$ Exposure

Examples of exposures for one participant during a calendar weekday during winter and summer are depicted in Figures 1 and 2, respectively. These time series plots represent the $PM_{2.5}$ exposures logged by the pDR in three minute intervals annotated with open text entries from the TADs. Figure 1 features a school day with the microenvironment 'at school' representing a baseline exposure of approximately 20 µg/m^3. This is notably higher than the other microenvironments encountered during the day. Also seen in this figure are short term peaks approximately 40 µg/m^3 while playing indoors and outdoors. The activity 'eating dinner' represents exposures above 100 µg/m^3. Figure 2 (a summer

day for the same participant) also features highly variable patterns in exposure. Exposures of 75 µg/m^3 are seen while in transit and at a restaurant. In both figures, baseline levels of $PM_{2.5}$ are seen to change by microenvironment and certain activities are associated with sharp 'peaks' or 'excursions' in $PM_{2.5}$ exposure. Table 2 presents the arithmetic and geometric average daily $PM_{2.5}$ exposures by season. It also presents percentages of time and exposure attributable to excursions in $PM_{2.5}$. Excursions are defined as levels that exceed the daily mean plus one, two and three standard deviations.

Microenvironmental times and $PM_{2.5}$ exposures

Descriptive statistics for daily values of personal $PM_{2.5}$ exposure as measured by the pDR, percent time, and percent exposure are presented in Table 3. Means of percent exposure received for each microenvironment are calculated only for those participants who spent time in those locations according to their TAD. The lowest exposures in both seasons occurred while participants were 'indoors at home'. As such, it was selected as the referent condition in the microenvironmental GEE models presented in Table 4. In winter, the children spent most of their time at home (67.1±12.7%) and at school (17.7±5.9%) where they received nearly all of their exposure (51.7±14.8% and 38.6±11.7%, respectively). One participant had no data representing time 'at school' as the two days of data were collected on a Saturday and a weekday statutory holiday.

GEE model estimates for winter revealed significant increases in $PM_{2.5}$ in all microenvironments except for 'outdoors at home' for which there was limited data. These increases were relatively equal in magnitude. In summer, the time no longer spent at school shifted to cause increases in times spent outdoors and indoors away from home. The substantial time spent 'indoors away from home' was noted as time spent in daycare, shopping and in restaurants. The outdoor environments represented a considerable percentage of exposure. Summer GEE model estimates of increases in $PM_{2.5}$ in microenvironments relative to 'indoors at home' revealed significant increases

in all microenvironments apart from 'in transit' and 'at work'. The microenvironment 'at work' was represented by one participant who spent ten hours in employment. 'Outdoors away from home' was associated with a mean increase of 10.3 μg/m^3 (95%CI: 6.6-13.9) of $PM_{2.5}$.

Personal activity times and $PM_{2.5}$ exposures

Daily means of percent exposure represented by each activity were only calculated for participants who performed them. Of the over 100 activity codes adopted from the NHAPS (Klepeis et al., 2001) and CHAPS (Leech et al., 1996) surveys, 70 were used in coding the personal activities reported by the participants. Of these, nearly 95% of the children's time was represented by 19 of the codes in winter and 17 in summer. The means of $PM_{2.5}$ pDR exposure, percent time, and percent of daily exposures for these activities are presented in Table 5. Night sleep represented the majority of the children's time in both seasons. It was observed that the median sleeping hours were 9PM-7AM and 10PM-9AM in the winter and summer, respectively (not reported). Table 6 presents the winter and summer GEE models for estimating increases in personal $PM_{2.5}$ while engaging in particle generating/resuspending activities while indoors at home. Of the 70 identified personal activities in the dataset, 32 were conducted indoors at home. These activities were predominantly sedentary (night sleep, watching TV, homework, etc.). The referent condition 'Sedentary' represented all sedentary personal activities other than 'Night sleep'. The exposure levels between these two categories were significantly different in both seasons with night sleep representing the lower $PM_{2.5}$ exposures, revealing a degree of diurnal variation in both seasons. While the children were indoors at home, the highest average increases in personal exposure were seen when they were involved in food preparation and indoor playing. Food preparation (which also included cooking) elevated average exposures by 40.7 μg/m^3 (95%CI 23.5-36.5) and 7.5 μg/m^3 (95%CI 1.6-13.4) in winter and summer respectively. 'Indoor playing' was reported by more of the participants and increased personal exposure on average by 10.1 μg/m^3 (95%CI 6.3-13.8) in the winter and 11.6 μg/m^3 (95%CI 8.1-15.5) in

the summer. 'Cleaning house' was associated with a significant average increase in exposure in winter only (9.6 μg/m^3 (95%CI 3.2-16.0)). Both in winter and summer, 'Eating' and 'personal hygiene' were reported by most participants and were associated with significant increases in their personal $PM_{2.5}$ exposure. 'Eating' was associated with average increases of 5.1 μg/m^3 (95%CI 3.0-7.1) and 3.0 μg/m^3 (95%CI 1.2-4.8) in winter and summer, respectively. This activity however does indicate a degree of reporting non-compliance by the participants as this activity was assuredly done by each of them every day. The GEE models representing data collected in both outdoor environments found no significant association between personal $PM_{2.5}$ exposure as measured by the pDR and personal activity. Therefore, no data on elevated levels of $PM_{2.5}$ by personal activity while outdoors are presented in this paper.

DISCUSSION

We found the microenvironmental profiles of the children in this study to be similar to time activity profiles reported in the U.S. National Human Activity Pattern Survey (NHAPS) and the Canadian Human Activity Pattern Survey (CHAPS) for respondents aged < 11 years (NHAPS n = 1,126; CHAPS n= 324) (Kleipis et al., 2001; Leech et al., 2002). For NHAPS and CHAPS, respectively, mean percent times spent 'indoors at home' (70.52±1.17, 72.33±2.38), 'outdoors' (4.2±0.5, 4.3±1.0), 'at school' (7.8±0.8, 5.7±1.2) and 'in vehicles' (3.6±0.3, 3.7±0.5) were comparable to our data when considering that their sampling methods included year round data collection and represented an oversampling of weekend days (Klepeis et al., 2001, Leech et al., 1996). With regards to the time spent in school, which was noted in the winter season only, the children spent an average of 67.1±12.7% of their time indoors at home and 17.7±5.9% of their time at school. These values are comparable to studies that have also monitored children during the school year (Noullett et al., 2006; Liu et al., 2003; Wu et al., 2005). Lastly, as the two seasonal sampling periods in this study were labelled as 'winter' and 'summer', they each also exclusively represented months where children were 'in school' and 'out

of school', respectively. As the impact of 'in school' on average daily $PM_{2.5}$ exposure was found to be substantial, it should be noted that it is a microenvironment occupied by children on a very consistent schedule ten months out of the year.

As can be seen in Table 3, the microenvironment 'indoors at home' had the lowest $PM_{2.5}$ concentration (ie. lowest exposure intensity); however, because participants spent most of their time there (67% winter, 72% summer), the greatest proportion of daily personal exposure occurred while indoors at home (52% winter, 66% summer). Comparatively, other microenvironments represented percentages of daily exposure much higher than those of time, revealing them to have much higher exposure intensities. Most notably, the outdoor microenvironments in both seasons had percentages of daily exposure 3 or 4 times higher than the percentages of time spent there. This situation may not be typical of most cities. In Wheeler et al., (2011b), the outdoor $PM_{2.5}$ levels were seen to exceed that of personal and indoor exposure and it was furthermore stated that this was not typical of most North American cities.

It's important to note that the microenvironments discussed in this paper can have different particle sources and with that represent different compositions and potential toxicities. Most notably, particulates of indoor and outdoor environments can represent different health risks. Outdoor environments represent particulates from combustion sources whereas indoor environments represent outdoor particulates that have infiltrated into the home and indoor sourced PM such as allergens and particulates resulting from personal activities. Estimates of the ambient fraction of indoor $PM_{2.5}$ exposure of the Windsor participants have been made using several infiltration methods (MacNeill et al., 2012). Indoor $PM_{2.5}$ exposure levels were 59% ambient in winter and 65% ambient in summer. Similarly, the increases in personal $PM_{2.5}$ associated with personal activities described in this paper can also represent particles of differing toxicities. The particles related to activities such as 'cleaning house' and 'indoor playing'

represent particle characteristics conceivably different than those of 'personal hygiene' and 'food preparation', for example.

The impact of various microenvironments encountered in a day on personal PM exposure has been studied in comparisons of direct personal exposure measures to estimates using microenvironmental (ME) modeling (Liu et al., 2003; Ozkaynak et al., 1996; Wu et al., 2005; Yip et al., 2004). These models sum up time-weighted exposures from various microenvironments to predict personal PM exposure. Regressing the ME estimates against directly measured personal exposure quantifies how much variability in the personal exposure data can be accounted for by the microenvironmental measures. In Liu et al., (2003), this approach was used for 133 elderly and 33 asthmatic children in Seattle, WA. $PM_{2.5}$ exposure data for personal and other microenvironments were measured using integrated 24h gravimetric sampling. The ME model estimates explained 46% to 65% percent of the variability in the elderly personal $PM_{2.5}$ exposure data and 9% of the children's. It was suggested that the lack of predictive power in the case of the children was due to their active lifestyle, as it was noted that the elderly subjects were typically more sedentary and had lower variability in the microenvironments they occupied. The use of integrated sampling in each microenvironment likely weakened the model's power as the microenvironmental exposure data reflected time when the participants were not present. Wu et al. (2005) applied the ME model for 20 asthmatic children in Alpine, CA, USA. These methods were improved by the use of continuous monitoring in each microenvironment. This allowed the use of microenvironmental exposure data specific to the times in which the participants occupied them to be used in the model. Hourly ME estimates were calculated and averaged into daily $PM_{2.5}$ means. When regressed against the directly measured personal exposure data, 6% and 48% of the variability was explained for the data representing children-days spent 'in-school' and 'not-in-school', respectively. This improved approach yielded a decent degree of explained variability in the case of the 'not-in-school' group. The low R^2 of the 'in-school' group was suggested to be due to

the use of exposure data collected in the 'indoors at home' microenvironment to represent time spent at school. The potential for difference in exposure between these two microenvironments is seen in our own microenvironmental analysis where we found $PM_{2.5}$ exposures at school to be 4.6 $\mu g/m^3$ (95%CI 3.0-6.2) higher than indoors at home and contribute an average of 38.6±11.7% of daily $PM_{2.5}$ exposure.

The use of GEE models to estimate differences in personal PM exposure measured with the pDR during particle generating activities and between microenvironments has been attempted elsewhere (Allen et al., 2004; Quintana et al., 2001). In Allen et al, (2004) 38 monitoring events (the monitoring of a subject for a single 5 or 10 day session) included healthy, elderly adults (n=5), elderly patients with congestive heart failure (n=11), or with chronic obstructive pulmonary disease (n=10) and asthmatic children (n=2). A significant increase of 7.88 $\mu g/m^3$ in PM exposure was seen for the microenvironment 'at work' which was represented by one participant. The microenvironment 'in transit' was a moderately significant (p=0.07) 1.62 $\mu g/m^3$ average increase in personal exposure. Particle generating activities 'at school' were associated with average increases of 5.76 $\mu g/m^3$ (p<0.001) and 8.3 $\mu g/m^3$ (p<0.001) in personal $PM_{2.5}$ during class and recess, respectively. Cooking was associated with an increase of 5.46 $\mu g/m^3$ (p<0.05) in personal exposure. Quintana et al, (2001) measured the personal PM exposure of ten non-smoking adult volunteers for one week using the pDR. Using the GEE method, $PM_{2.5}$ concentrations in microenvironments designated as 'outdoors' were, on average, 37.3 $\mu g/m^3$ higher than 'indoors' (the referent condition in their model). In their personal activity GEE model, activities such as yard work, construction, and cooking were also seen to have significantly elevated exposures.

In this paper, the methods of determining the effect of microenvironments and personal activities on elevated particle exposures proved effective. These methods included: the measurement of trends in $PM_{2.5}$ exposure with the pDR, the collection of activity pattern data with TADs and the analysis of these combined data using the GEE method.

However, some limitations are noted. The analysis was limited by an averaging time of 30 minutes imposed by the design of the TADs. This affects the analysis in several ways. Changes in activity and microenvironments can easily vary within 30 minute time periods. In Figure 1, a car commute is described in open text as a 'one minute' commute. The result of this entry would be the assignment of the activity 'car transit' to a 30 minute time period thereby over estimating its duration. Furthermore, the 30 minute average of the three minute pDR data assigned to 'car transit' would represent three minutes of true private car exposure and 27 minutes of misclassified exposure. In this particular case where the exposures before and after the commuting event are lower, we have an underestimate of exposure for the microenvironment 'in transit'. Other examples of peaks lasting less than 30 minutes can be seen in Figures 1 and 2 implying that the elimination of this error with the use of higher temporal resolution TAD data may result in refined exposure estimates for activities and microenvironments. Aside from the issue of misclassification, a reduction in variability results by averaging the three minute pDR data into 30 minute periods. Daily maximum values for each participant-day were reduced by a mean factor of 2.0 (range 1.1-6.5) when converting the 3 minute data to 30 minute averaging periods. Mean coefficients of variance (CV) for each season and participant combination were calculated and reduced from 164% (range 43-840%) to 143% (range 43-658%) when averaged to 30 minute periods. Given a finer temporal resolution for our data, this variability could be preserved. This is difficult to implement especially for children as compliance in completing TAD is challenging.

To monitor time activity information at higher temporal resolutions, novel approaches have begun to emerge. Use of Global positioning system (GPS) tracking devices show promise for more accurate reporting of time-location patterns of children aged 3-5 than parent completed diaries (Elgethun et al., 2007). The Personal Digital Assistant (PDA) system for Activity Registration and Recording of Travel Scheduling (PARROTS) collects both GPS and personal activity data using default answers and predefined dropdown lists. This method features a reduction in participant burden, an increase in temporal

resolution and has been found to decrease respondent error in comparison with paper diaries (Bellemans et al., 2008).

We combined time activity information collected with diaries and personal $PM_{2.5}$ exposure data collected with pDRs to relate personal exposure to microenvironments and activities. We found the effect of microenvironments and personal activities on elevated personal exposure to $PM_{2.5}$ to be significant. In winter, exposures while at school were found to be significantly elevated relative to indoors at home and constitute, on average, nearly 40% of the children's total daily exposure. We also found significant increases in personal $PM_{2.5}$ were attributed to personal activity while indoors at home. Although this panel study was conducted over a single year with a relatively small sample size of asthmatic children, who were not selected at random, their activity patterns are remarkably similar to those documented in national activity pattern surveys and other panel studies involving children. Understanding the sources of personal PM exposure and the significance of times spent near these sources is important when assigning exposure to populations and estimating the consequent health effects. Although centrally located fixed site monitoring data and microenvironmental models have some success with characterizing personal PM exposure, our findings indicate that the active lifestyle of children represents a significant factor in understanding their true $PM_{2.5}$ exposure.

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The authors would like to thank the reviewers for their considered review of the paper and their guidance on these improvements. We believe these improvements will help readers understand the paper.
We have included a response after each comment.

Reviewer: 1
Comments to the Author
A few minor revision points need to be resolved.

1) page 6, line 143. This sentence is redundant. A number of other sentences in the manuscript report the "three minute" averaging time.
Agreed. Sentence removed.

2) page 10, line 255. We are told here that there was a filter based PEM associated with the pDR. Earlier text indicated the PEM was "filterless". Were there two separate PEMs used in the backpack (one for the pDR and one for gravimetric analysis? Clarification is needed.
Sentence added into methods at line 143; "A second PEM was also included in the personal monitoring assembly." A second PEM, operating at 4.0 lpm, was also included in the personal monitoring assembly. This was a daily $PM_{2.5}$ sample which provided a gravimetric measure to which the pDR data could be compared."
Minor changes were also done to lines 259-261.

Reviewer: 2
Comments to the Author
Review of Manuscript ID JESEE-12-1494.R1

"Impact of microenvironments and personal activities on personal PM2.5 exposures among asthmatic children", by Van Ryswyk, Keith, Amanda Wheeler, Lance Wallace, Jill Kearney, Hongyu You, Ryan Kulka, and Iris Xiaohong Xu.

This manuscript makes an important contribution to the field by describing the microenvironment-resolved PM exposure of school-age children. The authors have been very responsive to reviewer comments, and the corresponding revisions have substantially improved the paper. These revisions include improved discussion of exposure intensity, the relationship between particle source and potential toxicity, outdoor exposures, and the influence of diurnal changes in activity pattern on exposure.

The manuscript is written clearly and reads well. I have only one editorial comment: on line 328, I suggest saying "significantly different" rather than "significant".
Agreed. Change made.