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



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Keywords:	personal exposure, particulate matter, inhalation exposure, exposure modeling, child exposure/health, analytical methods





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Journal of Exposure Analysis and Environmental Epidemiology





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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

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Season	N 3-min data	PM <sub>2.5</sub> GMDA	PM <sub>2.5</sub> MDA <sup>1</sup>	excursion = PM <sub>2.5</sub> > MDA+1SD		excursion = PM <sub>2.5</sub> > MDA+2SD		excursion = PM <sub>2.5</sub> > MDA+3SD	
	5 min data	µg/m <sup>3</sup>	µg/m <sup>3</sup>	% 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)

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Season	Microenvironment	Participants occupying microenvironment	n 30 minute periods	РМ <sub>2.5</sub> АМ (SD) (µg/m <sup>3</sup> )	ΡΜ <sub>2.5</sub> GM (GSD) (μg/m <sup>3</sup> )	Mean % Time (SD)	Mean % Daily Exposure (SD) <sup>1</sup>
	Indoors at Home	41	4688	8.9 (5.9)	6.4 (1.8)	67.1 (12.7)	51.7 (14.8)
Winter n=41	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)
	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)
Summer	In transit	31	230	19.3 (12.6)	13 (2.2)	3.5 (3.0)	8.9 (5.5)
n=35	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)

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	Winter (	n=41)					Summer	(n=34)				
Activity	N IDs	N 30min periods	AM(SD) PM <sub>2.5</sub> <sup>1</sup> (μg/m <sup>3</sup> )	GM(GSD) PM2.5 <sup>1</sup> (µg/m <sup>3</sup> )	% Time (SD)	% Daily Exposure (SD) <sup>2</sup>	N IDs	N 30min periods	AM(SD) PM2.5 <sup>1</sup>	GM(GSD) PM2.5 <sup>1</sup> (µg/m <sup>2</sup> )	% Time (SD)	% Daily Exposure (SD) <sup>2</sup>
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)	12(21)	14.2 (17.6)	20	192	12.7 (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)

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58 59 60 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 <sub>2.5</sub> (SE) (µg/m <sup>3</sup> )	Concentration difference from the referent condition (95% CI)
	Indoors at Home	41	4688	9.9 (0.8)	
	Outdoors at Home	16	49	10.9 (2.6)	0.9 (-3.0 - 4.9)
Winter	In transit	39	325	14.5 (1.3)	4.6 (3.2 - 6.0)**
n = 41	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)**
	Indoors at Home	35	4576	13.2 (1.3)	
	Outdoors at Home	29	284	16.4 (2.7)	3.2 (0.3 - 6.0)*
Summer	In transit	31	230	15.5 (1.9)	2.2 (-0.3 - 4.8)
n = 35	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)*

\*\*p<0.001, \*p<0.05

266x175mm (96 x 96 DPI)

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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 <sub>2.5</sub> (SE) (µg/m³)	Concentration difference from the referent condition (95% Cl)
Winter	Sedentary	41	954	10.7 (1.1)	
	Night sleep	41	3088	6.6 (0.7)	-4.1 (-5.72.4)**
	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 (-3.80.3)*
	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)

\*\*p<0.001, \*p<0.05

245x196mm (96 x 96 DPI)

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1 2		
3 4	1	Impact of microenvironments and personal activities on personal $PM_{2.5}$ exposures
5 6	2	among asthmatic children
7 8	3	
9 10	4	
10 11 12	5	Keith Van Ryswyk, BSc <sup>a</sup> , Amanda J. Wheeler, PhD <sup>a</sup> , Lance Wallace, PhD <sup>b</sup> , Jill Kearney,
13	6	MSc <sup>a</sup> , Hongyu You, MSc <sup>a</sup> , Ryan Kulka, BEng <sup>a</sup> , and Xiaohong Xu, PhD <sup>c</sup>
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16 17	8	
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# 30 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<sub>2.5</sub> 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<sub>2.5</sub> exposure. In summer, they spent 10.3±11.8% 'indoors away from home' which represented  $23.4 \pm 18.3\%$  of their PM<sub>2.5</sub> 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<sub>2.5</sub> exposures. Indoor playing represented a mean increase in  $PM_{2.5}$  of 10.1 µg/m<sup>3</sup> (95%Cl 6.3-13.8) and 11.6  $\mu$ g/m<sup>3</sup> (95%Cl 8.1-15.1) in winter and summer, respectively, as estimated by the pDR. 

50 Keywords: PM<sub>2.5</sub>; pDR; Personal exposure; Childhood asthma; personal activity

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# **CONFIDENTIAL MATERIAL**

# 53 INTRODUCTION

Several studies have found associations between fine particulate matter ( $PM_{25}$ ) 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<sub>2.5</sub> for a large area of a community. Community-wide estimates of ambient PM<sub>2.5</sub> can also be estimated using dispersion and land use regression modeling methods. However, studies comparing personal exposure levels of PM<sub>2.5</sub> 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.

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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

95 characterizes the effect of the children's personal activity patterns on their PM<sub>2.5</sub>

96 exposure by analyzing differences in personal PM<sub>2.5</sub> exposure by microenvironment and
 97 personal activity. An understanding of the effect of personal activity on personal PM<sub>2.5</sub>
 90 for a bit base of the bit base of the effect of personal activity on personal PM<sub>2.5</sub>

98 exposure for children can help identify the major sources and inform policy measures
99 designed to mitigate their exposure.

# 101 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

# **CONFIDENTIAL MATERIAL**

3	111	and ad the following Saturday afternoon. During each E day period, their percend DM
4 5	111	ended the following Saturday alternoon. During each 5 day period, their personal PM <sub>2.5</sub>
6	112	exposures were measured and information on their personal activity patterns collected.
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9 10	114	Time Activity Diary Data
11 12	115	Participants were asked to report their personal activities and microenvironmental
13	116	locations through the use of time activity diaries (TAD). For each 30 minute period in
14	117	their five days of monitoring, participants recorded their activity in open text and
16 17	118	indicated their location in one of six microenvironments. The six categories of
18 19	119	microenvironment were indoors at home, outdoors at home, in transit, at work/school,
20 21	120	outdoors away from home and indoors away from home. The category of 'at
22 23	121	work/school' was designated as such on account of the two seasons of the study. In
24 25	122	winter, this category was understood to represent time at school and in summer, time
26 27	123	spent in employment (one child reported working in the summer). TAD data was
28	124	entered into electronic form in duplicate and discrepancies resolved. Each personal
30	125	activity was classified using codes adapted from those of the National Human Activity
32	126	Pattern Survey (NHAPS) (Klepeis et al., 2001) and the Canadian Human Activity Pattern
33 34	127	Survey (CHAPS) (Leech et al., 1996). Coding was performed in duplicate to ensure
35 36	128	consistency in the categorization of the child's activity.
37 38	129	
39 40	130	Personal PM <sub>2.5</sub> Exposure Data
41 42	131	The personal DataRAM (pDR) (ThermoScientific, Waltham, MA, USA) has been
43 44	132	extensively used in the measurement of personal PM exposure (Quintana et al., 2001;
45 46	133	Wallace et al., 2003; Wallace et al., 2006; Wu et al., 2005; Yip et al., 2004). The pDR,
47	134	calibrated to a NIST particle standard, features a continuous and light weight method of
40 49	135	measuring particle concentrations in the air. It uses a laser at 880nm to measure mass
50 51	136	concentration. Each participant carried a pDR to continuously measure personal $PM_{2.5}$
52 53	137	over the five days using a three minute logging interval, to allow for the assignment of
54 55	138	personal activities and microenvironments identified through the TADs to the recorded
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exposure data. The pDR was also equipped with a Harvard Personal Environmental

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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<sub>2.5</sub> 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.

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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.

174 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:

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

179 where  $P_{kij}$  represents the percent contribution of category 'k' to the total exposure of 180 participant 'i' on day 'j',  $C_{kji}$  represents the average PM<sub>2.5</sub> concentration of category 'k' 181 for participant 'i' on day 'j', and  $F_{kji}$  represents the fraction of time spent in category 'k' 182 for participant 'i' on day 'j'. Arithmetic averages (across days) were calculated for 183 percent time, PM<sub>2.5</sub> exposure and percent exposure by season, participant and category. 184 These arithmetic means were used to calculate the arithmetic and geometric means by 185 season and category (across participants).

GEE models were used to estimate differences in personal PM<sub>2.5</sub> 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:

 $y = \beta_o + \sum \beta_i x_i + \varepsilon$  (Equation 2)

192 where y represents the exposure,  $\beta_0$  represents the model intercept, which is the 193 concentration of the referent condition in the model,  $\beta_i$  are the model coefficients,  $x_i$ 

194 represents the 0/1 indicator variables for the microenvironments or activities, and ε

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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<sub>i</sub> 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<sub>2.5</sub> 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

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2		
3 4	223	activity represents most night hours of each day when indoor exposures are typically
5 6	224	lower.
7 8	225	RESULTS
9 10	226	A total of 51 children were included in the 2006 WOEAS sampling sessions. Forty-eight
11 12	227	children diagnosed with asthma were recruited for the winter season with an additional
13	228	three recruited for the summer season on account of drop out. A total of 41 winter and
14	229	35 summer participants produced the required amount of pDR and TAD data to be
16	230	included in this analysis, 27 of which participated in both seasons. Most participants
18 19	231	lived in detached homes with forced air ventilation, were predominantly Caucasian and
20 21	232	between the ages of 10 and 13 (Table 1).
22 23	233	
24 25	234	Quality Assurance
26 27	235	In total, 204 598 3-minute personal $PM_{2.5}$ measures from 51 participants over the two
28 29	236	seasons were recorded in the study. Two sets of siblings were present in summer and
30 31	237	one in the winter. Measures belonging to one sibling of each pair were removed to
32	238	preserve the data's independence (n=8 704, 4.3%). Invalidation due to the end sample
33 34	239	flow varying by over 20% resulted in the loss of 11 551(5.6%) measures. Data loss on
35 36	240	account of pDR malfunction, including loss of battery power, pump failure and damaged
37 38	241	pDRs exhibiting unnatural trends in data, accounted for 36 932(18%) measures of
39 40	242	personal PM <sub>2.5</sub> . Positive drift corrections were performed on approximately 6% of the
41 42	243	data ranging from 2-6 $\mu$ g/m <sup>3</sup> . Negative drift corrections ranging from 2-4 $\mu$ g/m <sup>3</sup> were
43 44	244	applied to 0.3% of the records.
45 46	245	
47 48	246	The data was converted into 12 997 30-minute means for combination with the TAD
49	247	data. Minor remaining loss was accounted for by missing TAD data, the removal of all
50 51	248	30-minute periods during which field technicians were reported present by participants
52 53	249	and the data sufficiency criteria of at least seven 3-min pDR measures per 30-min mean,
54 55	250	18 hours of data per sample day and two days for every five day sampling session. The
56 57 58	251	resulting dataset included 12 873 30-minute periods from 41 and 35 participants in the

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winter and summer sessions, respectively. Of these participants, 27 were sampled inboth 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<sub>2.5</sub> measures. In comparison with the filter based PM<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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  $PM_{25}$  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<sup>3</sup>. 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<sup>3</sup> while playing indoors and outdoors. The activity 'eating dinner' represents exposures above 100 µg/m<sup>3</sup>. Figure 2 (a summer day for the same participant) also features highly variable patterns in exposure. Exposures of 75  $\mu$ g/m<sup>3</sup> are seen while in transit and at a restaurant. In both figures, 

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

288 Microenvironmental times and PM<sub>2.5</sub> exposures

289 Descriptive statistics for daily values of personal PM<sub>2.5</sub> exposure as measured by the 290 pDR, percent time, and percent exposure are presented in Table 3. Means of percent 291 exposure received for each microenvironment are calculated only for those participants 292 who spent time in those locations according to their TAD. The lowest exposures in both 293 seasons occurred while participants were 'indoors at home'. As such, it was selected as 294 the referent condition in the microenvironmental GEE models presented in Table 4. In 295 winter, the children spent most of their time at home (67.1±12.7%) and at school 296 (17.7±5.9%) where they received nearly all of their exposure (51.7±14.8% and 297 38.6±11.7%, respectively). One participant had no data representing time 'at school' as 298 the two days of data were collected on a Saturday and a weekday statutory holiday.

299

300 GEE model estimates for winter revealed significant increases in PM<sub>2.5</sub> in all 301 microenvironments except for 'outdoors at home' for which there was limited data. 302 These increases were relatively equal in magnitude. In summer, the time no longer 303 spent at school shifted to cause increases in times spent outdoors and indoors away 304 from home. The substantial time spent 'indoors away from home' was noted as time 305 spent in daycare, shopping and in restaurants. The outdoor environments represented 306 a considerable percentage of exposure. Summer GEE model estimates of increases in 307 PM<sub>2.5</sub> in microenvironments relative to 'indoors at home' revealed significant increases 308 in all microenvironments apart from 'in transit' and 'at work'. The microenvironment 'at 309 work' was represented by one participant who spent ten hours in employment.

310 'Outdoors away from home' was associated with a mean increase of 10.3 μg/m<sup>3</sup> (95%CI:
311 6.6-13.9) of PM<sub>2.5</sub>.

# 313 Personal activity times and PM<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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  $\mu$ g/m<sup>3</sup> (95%Cl 23.5-36.5) and 7.5 μg/m<sup>3</sup> (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  $\mu$ g/m<sup>3</sup> (95%Cl 6.3-13.8) in the winter and 11.6  $\mu$ g/m<sup>3</sup> (95%Cl 8.1-15.5) in the summer. 'Cleaning house' was associated with a significant average increase in exposure in winter only (9.6  $\mu$ g/m<sup>3</sup> (95%Cl 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<sub>2.5</sub> exposure. 'Eating' was associated with average increases of 5.1  $\mu$ g/m<sup>3</sup> (95%Cl 3.0-7.1) and 3.0  $\mu$ g/m<sup>3</sup> (95%Cl 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<sub>2.5</sub> exposure as measured by the pDR and personal activity. Therefore, no data on elevated levels of PM<sub>2.5</sub> 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'  $(3.6\pm0.3, 3.7\pm0.5)$  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<sub>2.5</sub> exposure 

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was found to be substantial, it should be noted that it is a microenvironment occupiedby 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<sub>2.5</sub> 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<sub>25</sub> exposure of the Windsor participants have been made using several infiltration methods (MacNeill et al., 2012). Indoor PM<sub>2.5</sub> exposure levels were 59% ambient in winter and 65% ambient in summer. Similarly, the increases in personal PM<sub>2.5</sub> 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.

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3 4	396	
5 6	397	The impact of various microenvironments encountered in a day on personal PM
7 8	398	exposure has been studied in comparisons of direct personal exposure measures to
9 10	399	estimates using microenvironmental (ME) modeling (Liu et al., 2003; Ozkaynak et al.,
10	400	1996; Wu et al., 2005; Yip et al., 2004). These models sum up time-weighted exposures
12 13	401	from various microenvironments to predict personal PM exposure. Regressing the ME
14 15	402	estimates against directly measured personal exposure quantifies how much variability
16 17	403	in the personal exposure data can be accounted for by the microenvironmental
18 19	404	measures. In Liu et al., (2003), this approach was used for 133 elderly and 33 asthmatic
20	405	children in Seattle, WA. PM <sub>2.5</sub> exposure data for personal and other microenvironments
22	406	were measured using integrated 24h gravimetric sampling. The ME model estimates
23	407	explained 46% to 65% percent of the variability in the elderly personal $PM_{2.5}$ exposure
25 26	408	data and 9% of the children's. It was suggested that the lack of predictive power in the
27 28	409	case of the children was due to their active lifestyle, as it was noted that the elderly
29 30	410	subjects were typically more sedentary and had lower variability in the
31 32	411	microenvironments they occupied. The use of integrated sampling in each
33 34	412	microenvironment likely weakened the model's power as the microenvironmental
35	413	exposure data reflected time when the participants were not present. Wu et al. (2005)
37	414	applied the ME model for 20 asthmatic children in Alpine, CA, USA. These methods
38 39	415	were improved by the use of continuous monitoring in each microenvironment. This
40 41	416	allowed the use of microenvironmental exposure data specific to the times in which the
42 43	417	participants occupied them to be used in the model. Hourly ME estimates were
44 45	418	calculated and averaged into daily $PM_{25}$ means. When regressed against the directly
46	419	measured personal exposure data. 6% and 48% of the variability was explained for the
48	420	data representing children-days spent (in-school' and (not-in-school' respectively. This
49 50	420	improved approach violded a decent degree of explained variability in the case of the
51 52	421	(not in school' group. The low $P^2$ of the (in school' group was suggested to be due to
53 54	422	the use of exposure data collected in the 'indepersat home' microanvironment to
55 56	423	the use of exposure data collected in the indoors at nome microenvironment to
57 57	424	represent time spent at school. The potential for difference in exposure between these

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two microenvironments is seen in our own microenvironmental analysis where we found PM<sub>2.5</sub> exposures at school to be 4.6 μg/m<sup>3</sup> (95%Cl 3.0-6.2) higher than indoors at home and contribute an average of 38.6±11.7% of daily PM<sub>2.5</sub> 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<sup>3</sup> 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<sup>3</sup> 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<sub>2.5</sub> during class and recess, respectively. Cooking was associated with an increase of 5.46  $\mu$ g/m<sup>3</sup> (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<sub>2.5</sub> 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.

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454 Changes in activity and microenvironments can easily vary within 30 minute time 455 periods. In Figure 1, a car commute is described in open text as a 'one minute' 456 commute. The result of this entry would be the assignment of the activity 'car transit' to 457 a 30 minute time period thereby over estimating its duration. Furthermore, the 30 458 minute average of the three minute pDR data assigned to 'car transit' would represent 459 three minutes of true private car exposure and 27 minutes of misclassified exposure. In 460 this particular case where the exposures before and after the commuting event are 461 lower, we have an underestimate of exposure for the microenvironment 'in transit'. 462 Other examples of peaks lasting less than 30 minutes can be seen in Figures 1 and 2 463 implying that the elimination of this error with the use of higher temporal resolution 464 TAD data may result in refined exposure estimates for activities and microenvironments. 465 Aside from the issue of misclassification, a reduction in variability results by averaging 466 the three minute pDR data into 30 minute periods. Daily maximum values for each 467 participant-day were reduced by a mean factor of 2.0 (range 1.1-6.5) when converting 468 the 3 minute data to 30 minute averaging periods. Mean coefficients of variance (CV) 469 for each season and participant combination were calculated and reduced from 164% 470 (range 43-840%) to 143% (range 43-658%) when averaged to 30 minute periods. Given a 471 finer temporal resolution for our data, this variability could be preserved. This is difficult 472 to implement especially for children as compliance in completing TAD is challenging. 473

474 To monitor time activity information at higher temporal resolutions, novel approaches 475 have begun to emerge. Use of Global positioning system (GPS) tracking devices show 476 promise for more accurate reporting of time-location patterns of children aged 3-5 than 477 parent completed diaries (Elgethun et al., 2007). The Personal Digital Assistant (PDA) 478 system for Activity Registration and Recording of Travel Scheduling (PARROTS) collects 479 both GPS and personal activity data using default answers and predefined dropdown 480 lists. This method features a reduction in participant burden, an increase in temporal 481 resolution and has been found to decrease respondent error in comparison with paper 482 diaries (Bellemans et al., 2008).

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483	
484	We combined time activity information collected with diaries and personal $PM_{2.5}$
485	exposure data collected with pDRs to relate personal exposure to microenvironments
486	and activities. We found the effect of microenvironments and personal activities on
487	elevated personal exposure to $PM_{2.5}$ to be significant. In winter, exposures while at
488	school were found to be significantly elevated relative to indoors at home and
489	constitute, on average, nearly 40% of the children's total daily exposure. We also found
490	significant increases in personal PM <sub>2.5</sub> were attributed to personal activity while indoors
491	at home. Although this panel study was conducted over a single year with a relatively
492	small sample size of asthmatic children, who were not selected at random, their activity
493	patterns are remarkably similar to those documented in national activity pattern
494	surveys and other panel studies involving children. Understanding the sources of
495	personal PM exposure and the significance of times spent near these sources is
496	important when assigning exposure to populations and estimating the consequent
497	health effects. Although centrally located fixed site monitoring data and
498	microenvironmental models have some success with characterizing personal PM
499	exposure, our findings indicate that the active lifestyle of children represents a
500	significant factor in understanding their true $PM_{2.5}$ exposure.
501	
502	ACKNOWLEDGEMENTS
503	We would like to thank the study participants, field technicians and Nina Dobbin (Clark)
504	and Morgan MacNeill for conducting the internal review. Funding for this work was
505	provided by the Border Air Quality Strategy.
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8 9	1	Impact of microenvironments and personal activities on personal PM <sub>2.5</sub> exposures
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14 15	5	Keith Van Ryswyk, BSc <sup>a</sup> , Amanda J. Wheeler, PhD <sup>a</sup> , Lance Wallace, PhD <sup>b</sup> , Jill Kearney,
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#### 30 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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> exposure. In summer, they spent 10.3±11.8% 'indoors away from home' which represented 23.4±18.3% of their PM<sub>2.5</sub> 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<sub>25</sub> exposures. Indoor playing represented a mean increase in  $PM_{2.5}$  of 10.1 µg/m<sup>3</sup> (95%Cl 6.3-13.8) and 11.6  $\mu$ g/m<sup>3</sup> (95%Cl 8.1-15.1) in winter and summer, respectively, as estimated by the pDR. Keywords: PM<sub>2.5</sub>; pDR; Personal exposure; Childhood asthma; personal activity 

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INTRODUCTION

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67 stress of air pollution impedes the process of pulmonary morphogenesis during 68 childhood, resulting in decreased lung function that impacts quality of life in adulthood 69 and old age (Gauderman et al., 2004). 70 Fixed-site monitor (FSM) data has been used extensively as a surrogate for personal 71 72 exposure to ambient pollution in many air pollution health effect studies (Dales et al., 73 2008; Dockery, 2001; Gauderman et al., 2004; Li et al., 2011). In the case of PM<sub>2.5</sub>, its 74 regionally consistent nature allows for the assumption that the concentrations 75 measured at a single FSM may be used as a relevant measure of ambient  $PM_{2.5}$  for a 76 large area of a community. Community-wide estimates of ambient  $PM_{2.5}$  can also be 77 estimated using dispersion and land use regression modeling methods. However, 78 studies comparing personal exposure levels of PM<sub>2.5</sub> to that of FSMs often cite a wide 79 variation of correlations across participants (Adgate et al., 2003; Brown et al., 2009; Crist 80 et al., 2008; Wallace, 2000). In many of these comparisons, personal activity and 81 exposure to indoor sources has been suggested as potential sources of the variability.

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.,

et al., 2010; Kulkarni and Grigg, 2008; O'Connor et al., 2008). In contrast to adults,

1999; Strickland et al., 2010), and decreased lung function (Gauderman et al., 2004; He

children represent an especially sensitive population to PM<sub>2.5</sub> 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

82	Personal PM exposures have also been compared to estimates of exposure from
83	microenvironmental (ME) modeling (Liu et al., 2003; Ozkaynak et al., 1996; Wu et al.,
84	2005; Yip et al., 2004). These studies demonstrated that microenvironment-specific
85	exposure data can account for a good deal of personal PM variability in elderly
86	populations where most of their time is spent at home and where sedentary lifestyles
87	are common. Considerable unexplained variability remains with pediatric subjects
88	alluding to the question of what effects personal activities and the different
89	microenvironments of children have on their personal PM exposure.
90	
91	The Windsor, Ontario Exposure Assessment Study (WOEAS) involved the monitoring of
92	48 asthmatic children for 5 consecutive days in both the winter and summer of 2006.
93	Personal activities and times spent in various microenvironments were recorded in time
94	activity diaries and personal $PM_{2.5}$ exposure was monitored using the pDR. This paper
95	characterizes the effect of the children's personal activity patterns on their $PM_{2.5}$
96	exposure by analyzing differences in personal $PM_{2.5}$ exposure by microenvironment and
97	personal activitiy. An understanding of the effect of personal activity on personal PM $_{ m 2.5}$
98	exposure for children can help identify the major sources and inform policy measures
99	designed to mitigate their exposure.
100	
101	METHODS
102	The methods used in WOEAS are more fully documented elsewhere (Wallace et al.,
103	2011; Wheeler et al., 2011a). Briefly, participants were selected from recruits of the
104	Windsor Children's Respiratory Health study (Dales et al., 2008). Using information from
105	that study, children between 10-13 years with doctor diagnosed asthma living in non-
106	smoking residences were recruited for the WOEAS in both the winter and summer of
107	2006. In each season, 48 children were monitored for a period of five consecutive days,
108	from Monday to Saturday; technicians visited the participants daily. Each season, six
109	participants were monitored concurrently during each of the eight sampling weeks.
110	Each five day sampling week began on a Monday evening at approximately 4PM and

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8 9	111	ended the following Saturday afternoon. During each 5 day period, their personal $PM_{2.5}$
10	112	exposures were measured and information on their personal activity patterns collected.
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13	114	Time Activity Diary Data
14 15	115	Participants were asked to report their personal activities and microenvironmental
16	116	locations through the use of time activity diaries (TAD). For each 30 minute period in
17	117	their five days of monitoring, participants recorded their activity in open text and
19 20	118	indicated their location in one of six microenvironments. The six categories of
20	119	microenvironment were indoors at home, outdoors at home, in transit, at work/school,
22 23 24 25 26 27 28 29 30 31 32 33 34 35	120	outdoors away from home and indoors away from home. The category of 'at
	121	work/school' was designated as such on account of the two seasons of the study. In
	122	winter, this category was understood to represent time at school and in summer, time
	123	spent in employment (one child reported working in the summer). TAD data was
	124	entered into electronic form in duplicate and discrepancies resolved. Each personal
	125	activity was classified using codes adapted from those of the National Human Activity
	126	Pattern Survey (NHAPS) (Klepeis et al., 2001) and the Canadian Human Activity Pattern
	127	Survey (CHAPS) (Leech et al., 1996). Coding was performed in duplicate to ensure
	128	consistency in the categorization of the child's activity.
36 37	129	
38	130	Personal PM <sub>2.5</sub> Exposure Data
39 40	131	The personal DataRAM (pDR) (ThermoScientific, Waltham, MA, USA) has been
41	132	extensively used in the measurement of personal PM exposure (Quintana et al., 2001;
42 43	133	Wallace et al., 2003; Wallace et al., 2006; Wu et al., 2005; Yip et al., 2004). The pDR,
44 45	134	calibrated to a NIST particle standard, features a continuous and light weight method of
45 46	135	measuring particle concentrations in the air. It uses a laser at 880nm to measure mass
47	136	concentration. Each participant carried a pDR to continuously measure personal PM <sub>2.5</sub>
49	137	over the five days using a three minute logging interval, to allow for the assignment of
50 51	138	personal activities and microenvironments identified through the TADs to the recorded
52	139	exposure data. The pDR was also equipped with a Harvard <del>pP</del> ersonal eEnvironmental
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mMonitor (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<sub>2.5</sub> sample which provided a gravimetric measure to which the pDR data could be compared. Concentrations were logged in three minute intervals. 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. 

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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.

#### 175 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:

179 
$$P_{kij} = \frac{C_{kij} \times F_{kij}}{\sum_{k=1}^{n} C_{kij} \times F_{kij}}$$
 (Equation 1)

180 where  $P_{kij}$  represents the percent contribution of category 'k' to the total exposure of 181 participant 'i' on day 'j',  $C_{kji}$  represents the average PM<sub>2.5</sub> concentration of category 'k' 182 for participant 'i' on day 'j', and  $F_{kji}$  represents the fraction of time spent in category 'k' 183 for participant 'i' on day 'j'. Arithmetic averages (across days) were calculated for 184 percent time, PM<sub>2.5</sub> exposure and percent exposure by season, participant and category. 185 These arithmetic means were used to calculate the arithmetic and geometric means by 186 season and category (across participants). 

GEE models were used to estimate differences in personal PM<sub>2.5</sub> 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:

$$y = \beta_o + \sum \beta_i x_i + \varepsilon$$
 (Equation 2)

where y represents the exposure, β<sub>o</sub> represents the model intercept, which is the
 concentration of the referent condition in the model, β<sub>i</sub> are the model coefficients, x<sub>i</sub>
 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<sub>i</sub> 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<sub>i</sub>, 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<sub>2.5</sub> 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

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8 9	224	activity represents most night hours of each day when indoor exposures are typically
10	225	lower.
11 12	226	RESULTS
13	227	A total of 51 children were included in the 2006 WOEAS sampling sessions. Forty-eight
14 15	228	children diagnosed with asthma were recruited for the winter season with an additional
16	229	three recruited for the summer season on account of drop out. A total of 41 winter and
17 18	230	35 summer participants produced the required amount of pDR and TAD data to be
19	231	included in this analysis, 27 of which participated in both seasons. Most participants
20 21	232	lived in detached homes with forced air ventilation, were predominantly Caucasian and
22	233	between the ages of 10 and 13 (Table 1).
23 24	234	
25 26 27 28 29 30 31 32 33	235	Quality Assurance
	236	In total, 204 598 3-minute personal PM $_{25}$ measures from 51 participants over the two
	237	seasons were recorded in the study. Two sets of siblings were present in summer and
	238	one in the winter. Measures belonging to one sibling of each pair were removed to
	239	preserve the data's independence (n=8 704, 4.3%). Invalidation due to the end sample
	240	flow varying by over 20% resulted in the loss of 11 551(5.6%) measures. Data loss on
34 35	241	account of pDR malfunction, including loss of battery power, pump failure and damaged
36	242	pDRs exhibiting unnatural trends in data, accounted for 36 932(18%) measures of
37 38	243	personal $PM_{25}$ . Positive drift corrections were performed on approximately 6% of the
39	244	data ranging from 2-6 $\mu$ g/m <sup>3</sup> . Negative drift corrections ranging from 2-4 $\mu$ g/m <sup>3</sup> were
40 41	245	applied to 0.3% of the records.
42 42	246	
43 44	247	The data was converted into 12 997 30-minute means for combination with the TAD
45 46	248	data Minor remaining loss was accounted for by missing TAD data, the removal of all
40	249	30-minute periods during which field technicians were reported present by participants
48 40	250	and the data sufficiency criteria of at least seven 3-min pDR measures per 30-min mean.
50	251	18 hours of data per sample day and two days for every five day sampling session. The
51 52	252	resulting dataset included 12 873 30-minute periods from 41 and 35 participants in the
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42 43 44 45 46 47 48 49 50 51 52 53 54 55 56 57	246 247 248 249 250 251 252	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

233	winter and summer sessions, respectively. Of these participants, 27 were sampled in
254	both seasons. The number of days per participant ranged from two to five.
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256	The performance of the pDR-1000 in this study is documented in Wallace et al, (2011)
257	Due to its dependence on the index of refraction and the density of the aerosol mixtu
258	(Görner et al., 1995), the exposures measured by this unit are typically overestimated
259	compared to gravimetric PM <sub>2.5</sub> measures. Along with the pDR, each participant carrie
260	daily PM <sub>2.5</sub> -personal environment monitors (PEM; Chempass System R&P/Thermo) Ir
261	comparison with these filter based $PM_{2.5}$ <u>PEM</u> data, the pDR over predicted $PM_{2.5}$ by
262	60% with an R <sup>2</sup> of 71.4% (Wallace et al., 2011). Each data point in this comparison
263	represents 24 hours of personal $PM_{2.5}$ exposure, each representing a unique
264	combination of time spent in various microenvironments. As different
265	microenvironments have varying levels of relative humidity and particle properties,
266	which can affect the relationship between these two methods of measuring $PM_{2.5}$ , it i
267	reassuring to see this consistent relationship without having adjusted for this factor. T
268	data presented in this paper have not been adjusted to the gravimetric method as thi
269	bias does not affect the relative relationships of measurements in different
270	microenvironments and personal activities. It should be noted that these levels are n
271	to be interpreted as actual PM <sub>2.5</sub> exposure values.
272	
273	3-min PM <sub>2.5</sub> Exposure
274	Examples of exposures for one participant during a calendar weekday during winter a
275	summer are depicted in Figures 1 and 2, respectively. These time series plots represent
276	the $PM_{2.5}$ exposures logged by the pDR in three minute intervals annotated with oper
277	text entries from the TADs. Figure 1 features a school day with the microenvironment
278	'at school' representing a baseline exposure of approximately 20 $\mu\text{g/m}^3.$ This is notable
279	higher than the other microenvironments encountered during the day. Also seen in th
280	figure are short term peaks approximately 40 $\mu$ g/m <sup>3</sup> while playing indoors and outdoor
281	The activity 'eating dinner' represents exposures above 100 $\mu$ g/m <sup>3</sup> . Figure 2 (a summe

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8	282	day for the same participant) also features highly variable patterns in exposure.
9 10	283	Exposures of 75 $\mu$ g/m <sup>3</sup> are seen while in transit and at a restaurant. In both figures,
11 12	284	baseline levels of PM <sub>2.5</sub> are seen to change by microenvironment and certain activities
13	285	are associated with sharp 'peaks' or 'excursions' in PM <sub>2.5</sub> exposure. Table 2 presents the
14 15 16 17 18 19 20 21 22 23 24 25 26	286	arithmetic and geometric average daily PM <sub>2.5</sub> exposures by season. It also presents
	287	percentages of time and exposure attributable to excursions in PM <sub>2.5</sub> . Excursions are
	288	defined as levels that exceed the daily mean plus one, two and three standard
	289	deviations.
	290	
	291	Microenvironmental times and PM <sub>2.5</sub> exposures
	292	Descriptive statistics for daily values of personal $PM_{2.5}$ exposure as measured by the
	293	pDR, percent time, and percent exposure are presented in Table 3. Means of percent
27	294	exposure received for each microenvironment are calculated only for those participants
28 29 30 31 32 33 34 35 36 37 38	295	who spent time in those locations according to their TAD. The lowest exposures in both
	296	seasons occurred while participants were 'indoors at home'. As such, it was selected as
	297	the referent condition in the microenvironmental GEE models presented in Table 4. In
	298	winter, the children spent most of their time at home (67.1±12.7%) and at school
	299	(17.7±5.9%) where they received nearly all of their exposure (51.7±14.8% and
	300	38.6±11.7%, respectively). One participant had no data representing time 'at school' as
	301	the two days of data were collected on a Saturday and a weekday statutory holiday.
39 40	302	
41	303	GEE model estimates for winter revealed significant increases in PM <sub>2.5</sub> in all
42 43	304	microenvironments except for 'outdoors at home' for which there was limited data.
44 45 46	305	These increases were relatively equal in magnitude. In summer, the time no longer
	306	spent at school shifted to cause increases in times spent outdoors and indoors away
47 49	307	from home. The substantial time spent 'indoors away from home' was noted as time
48 49	308	spent in daycare, shopping and in restaurants. The outdoor environments represented
50 51	309	a considerable percentage of exposure. Summer GEE model estimates of increases in
52 53 54	310	$\ensuremath{PM_{2.5}}$ 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  $10.3 \,\mu\text{g/m}^3$  (95%CI: 6.6-13.9) of PM<sub>2.5</sub>. Personal activity times and PM<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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  $\mu$ g/m<sup>3</sup> (95%Cl 23.5-36.5) and 7.5 μg/m<sup>3</sup> (95%Cl 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  $\mu$ g/m<sup>3</sup> (95%Cl 6.3-13.8) in the winter and 11.6  $\mu$ g/m<sup>3</sup> (95%Cl 8.1-15.5) in

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7 8	340	the summer 'Cleaning house' was associated with a significant average increase in
9	241	the summer. Cleaning house was associated with a significant average increase in
10	341	exposure in winter only (9.6 μg/m (95%ci 3.2-16.0)). Both in winter and summer,
12	342	'Eating' and 'personal hygiene' were reported by most participants and were associated
13 14	343	with significant increases in their personal PM <sub>2.5</sub> exposure. 'Eating' was associated with
15	344	average increases of 5.1 $\mu$ g/m <sup>3</sup> (95%Cl 3.0-7.1) and 3.0 $\mu$ g/m <sup>3</sup> (95%Cl 1.2-4.8) in winter
16	345	and summer, respectively. This activity however does indicate a degree of reporting
18	346	non-compliance by the participants as this activity was assuredly done by each of them
19	347	every day. The GEE models representing data collected in both outdoor environments
20 21	348	found no significant association between personal PM <sub>2.5</sub> exposure as measured by the
22	349	pDR and personal activity. Therefore, no data on elevated levels of PM <sub>2.5</sub> by personal
23 24	350	activity while outdoors are presented in this paper.
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20 27	352	DISCUSSION
28 29	353	
30	354	We found the microenvironmental profiles of the children in this study to be similar to
31 32	355	time activity profiles reported in the U.S. National Human Activity Pattern Survey
33	356	(NHAPS) and the Canadian Human Activity Pattern Survey (CHAPS) for respondents aged
34 35	357	< 11 years (NHAPS n = 1,126; CHAPS n= 324) (Kleipis et al., 2001; Leech et al., 2002). For
36 37	358	NHAPS and CHAPS, respectively, mean percent times spent 'indoors at home'
38	359	(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
39 40	360	'in vehicles' (3.6 $\pm$ 0.3, 3.7 $\pm$ 0.5) were comparable to our data when considering that their
41	361	sampling methods included year round data collection and represented an oversampling
42 43	362	of weekend days (Klepeis et al., 2001, Leech et al., 1996). With regards to the time
44	363	spent in school, which was noted in the winter season only, the children spent an
45 46	364	average of 67.1±12.7% of their time indoors at home and 17.7±5.9% of their time at
47	365	school. These values are comparable to studies that have also monitored children
48 49	366	during the school year (Noullett et al., 2006; Liu et al., 2003; Wu et al., 2005). Lastly, as
50	367	the two seasonal sampling periods in this study were labelled as 'winter' and 'summer',
51 52	368	they each also exclusively represented months where children were 'in school' and 'out
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369 370 371 372	of school', respectively. As the impact of 'in school' on average daily PM <sub>2.5</sub> 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 <sub>2.5</sub> 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,
370 371 372	<ul> <li>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.</li> <li>As can be seen in Table 3, the microenvironment 'indoors at home' had the lowest PM<sub>2.5</sub> 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,</li> </ul>
371 372	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 <sub>2.5</sub> 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,
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374	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,
375	exposure occurred while indoors at home (52% winter, 66% summer). Comparatively,
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377	other microenvironments represented percentages of daily exposure much higher than
378	those of time, revealing them to have much higher exposure intensities. Most notably,
379	the outdoor microenvironments in both seasons had percentages of daily exposure 3 or
380	4 times higher than the percentages of time spent there. This situation may not be
381	typical of most cities. In Wheeler et al., (2011b), the outdoor $PM_{2.5}$ levels were seen to
382	exceed that of personal and indoor exposure and it was furthermore stated that this
383	was not typical of most North American cities.
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385	It's important to note that the microenvironments discussed in this paper can have
386	different particle sources and with that represent different compositions and potential
387	toxicities. Most notably, particulates of indoor and outdoor environments can
388	represent different health risks. Outdoor environments represent particulates from
389	combustion sources whereas indoor environments represent outdoor particulates that
390	have infiltrated into the home and indoor sourced PM such as allergens and particulates
391	resulting from personal activities. Estimates of the ambient fraction of indoor PM <sub>2.5</sub>
392	exposure of the Windsor participants have been made using several infiltration methods
393	(MacNeill et al., 2012). Indoor PM $_{2.5}$ exposure levels were 59% ambient in winter and
394	65% ambient in summer. Similarly, the increases in personal $PM_{2.5}$ associated with
395	personal activities described in this paper can also represent particles of differing
396	toxicities. The particles related to activities such as 'cleaning house' and 'indoor playing'

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9 10 11	397	represent particle characteristics conceivably different than those of 'personal hygiene'
	398	and 'food preparation', for example.
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13 14	400	The impact of various microenvironments encountered in a day on personal PM
15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36	401	exposure has been studied in comparisons of direct personal exposure measures to
	402	estimates using microenvironmental (ME) modeling (Liu et al., 2003; Ozkaynak et al.,
	403	1996; Wu et al., 2005; Yip et al., 2004). These models sum up time-weighted exposures
	404	from various microenvironments to predict personal PM exposure. Regressing the ME
	405	estimates against directly measured personal exposure quantifies how much variability
	406	in the personal exposure data can be accounted for by the microenvironmental
	407	measures. In Liu et al., (2003), this approach was used for 133 elderly and 33 asthmatic
	408	children in Seattle, WA. PM <sub>2.5</sub> exposure data for personal and other microenvironments
	409	were measured using integrated 24h gravimetric sampling. The ME model estimates
	410	explained 46% to 65% percent of the variability in the elderly personal PM <sub>2.5</sub> exposure
	411	data and 9% of the children's. It was suggested that the lack of predictive power in the
	412	case of the children was due to their active lifestyle, as it was noted that the elderly
	413	subjects were typically more sedentary and had lower variability in the
	414	microenvironments they occupied. The use of integrated sampling in each
	415	microenvironment likely weakened the model's power as the microenvironmental
37 38	416	exposure data reflected time when the participants were not present. Wu et al. (2005)
39	417	applied the ME model for 20 asthmatic children in Alpine, CA, USA. These methods
40 41	418	were improved by the use of continuous monitoring in each microenvironment. This
42	419	allowed the use of microenvironmental exposure data specific to the times in which the
43 44 45 46 47 48 49 50	420	participants occupied them to be used in the model. Hourly ME estimates were
	421	calculated and averaged into daily PM <sub>2.5</sub> means. When regressed against the directly
	422	measured personal exposure data, 6% and 48% of the variability was explained for the
	423	data representing children-days spent 'in-school' and 'not-in-school', respectively. This
	424	improved approach yielded a decent degree of explained variability in the case of the
51 52	425	'not-in-school' group. The low $R^2$ of the 'in-school' group was suggested to be due to
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426	the use of exposure data collected in the 'indoors at home' microenvironment to
427	represent time spent at school. The potential for difference in exposure between these
428	two microenvironments is seen in our own microenvironmental analysis where we
429	found $PM_{2.5}$ exposures at school to be 4.6 $\mu$ g/m <sup>3</sup> (95%Cl 3.0-6.2) higher than indoors at
430	home and contribute an average of $38.6\pm11.7\%$ of daily PM <sub>2.5</sub> exposure.
431	
432	The use of GEE models to estimate differences in personal PM exposure measured with
433	the pDR during particle generating activities and between microenvironments has been
434	attempted elsewhere (Allen et al., 2004; Quintana et al., 2001). In Allen et al, (2004) 38
435	monitoring events (the monitoring of a subject for a single 5 or 10 day session) included
436	healthy, elderly adults (n=5), elderly patients with congestive heart failure (n=11), or
437	with chronic obstructive pulmonary disease (n=10) and asthmatic children (n=2). A
438	significant increase of 7.88 $\mu$ g/m <sup>3</sup> in PM exposure was seen for the microenvironment
439	'at work' which was represented by one participant. The microenvironment 'in transit'
440	was a moderately significant ( $p$ =0.07) 1.62 $\mu$ g/m <sup>3</sup> average increase in personal exposure
441	Particle generating activities 'at school' were associated with average increases of 5.76
442	$\mu$ g/m <sup>3</sup> ( $p$ <0.001) and 8.3 $\mu$ g/m <sup>3</sup> ( $p$ <0.001) in personal PM <sub>2.5</sub> during class and recess,
443	respectively. Cooking was associated with an increase of 5.46 $\mu$ g/m <sup>3</sup> (p<0.05) in
444	personal exposure. Quintana et al, (2001) measured the personal PM exposure of ten
445	non-smoking adult volunteers for one week using the pDR. Using the GEE method, $\mathrm{PM}_{2.5}$
446	concentrations in microenvironments designated as 'outdoors' were, on average, 37.3
447	$\mu$ g/m <sup>3</sup> higher than 'indoors' (the referent condition in their model). In their personal
448	activity GEE model, activities such as yard work, construction, and cooking were also
449	seen to have significantly elevated exposures.
450	
451	In this paper, the methods of determining the effect of microenvironments and persona
452	activities on elevated particle exposures proved effective. These methods included: the
453	measurement of trends in $PM_{2.5}$ exposure with the pDR, the collection of activity
454	pattern data with TADs and the analysis of these combined data using the GEE method.
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8	455	However, some limitations are noted. The analysis was limited by an averaging time of
10	456	30 minutes imposed by the design of the TADs. This affects the analysis in several ways.
11 12 13 14 15 16	457	Changes in activity and microenvironments can easily vary within 30 minute time
	458	periods. In Figure 1, a car commute is described in open text as a 'one minute'
	459	commute. The result of this entry would be the assignment of the activity 'car transit' to
	460	a 30 minute time period thereby over estimating its duration. Furthermore, the 30
17 18	461	minute average of the three minute pDR data assigned to 'car transit' would represent
19	462	three minutes of true private car exposure and 27 minutes of misclassified exposure. In
20 21	463	this particular case where the exposures before and after the commuting event are
22	464	lower, we have an underestimate of exposure for the microenvironment 'in transit'.
23 24 25	465	Other examples of peaks lasting less than 30 minutes can be seen in Figures 1 and 2
	466	implying that the elimination of this error with the use of higher temporal resolution
26 27	467	TAD data may result in refined exposure estimates for activities and microenvironments.
28 29 30 31 32 33	468	Aside from the issue of misclassification, a reduction in variability results by averaging
	469	the three minute pDR data into 30 minute periods. Daily maximum values for each
	470	participant-day were reduced by a mean factor of 2.0 (range 1.1-6.5) when converting
	471	the 3 minute data to 30 minute averaging periods. Mean coefficients of variance (CV)
34 35	472	for each season and participant combination were calculated and reduced from 164%
36	473	(range 43-840%) to 143% (range 43-658%) when averaged to 30 minute periods. Given a
37 38	474	finer temporal resolution for our data, this variability could be preserved. This is difficult
39	475	to implement especially for children as compliance in completing TAD is challenging.
40 41	476	
42	477	To monitor time activity information at higher temporal resolutions, novel approaches
43 44	478	have begun to emerge. Use of Global positioning system (GPS) tracking devices show
45 46 47 48 49 50	479	promise for more accurate reporting of time-location patterns of children aged 3-5 than
	480	parent completed diaries (Elgethun et al., 2007). The Personal Digital Assistant (PDA)
	481	system for Activity Registration and Recording of Travel Scheduling (PARROTS) collects
	482	both GPS and personal activity data using default answers and predefined dropdown
51	483	lists. This method features a reduction in participant burden, an increase in temporal
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484	resolution and has been found to decrease respondent error in comparison with paper
485	diaries (Bellemans et al., 2008).
486	
487	We combined time activity information collected with diaries and personal $PM_{2.5}$
488	exposure data collected with pDRs to relate personal exposure to microenvironments
489	and activities. We found the effect of microenvironments and personal activities on
490	elevated personal exposure to $PM_{2.5}$ to be significant. In winter, exposures while at
491	school were found to be significantly elevated relative to indoors at home and
492	constitute, on average, nearly 40% of the children's total daily exposure. We also found
493	significant increases in personal PM2.5 were attributed to personal activity while indoors
494	at home. Although this panel study was conducted over a single year with a relatively
495	small sample size of asthmatic children, who were not selected at random, their activity
496	patterns are remarkably similar to those documented in national activity pattern
497	surveys and other panel studies involving children. Understanding the sources of
498	personal PM exposure and the significance of times spent near these sources is
499	important when assigning exposure to populations and estimating the consequent
500	health effects. Although centrally located fixed site monitoring data and
501	microenvironmental models have some success with characterizing personal PM
502	exposure, our findings indicate that the active lifestyle of children represents a
503	significant factor in understanding their true PM <sub>2.5</sub> exposure.
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505	ACKNOWLEDGEMENTS
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507	and Morgan MacNeill for conducting the internal review. Funding for this work was
508	provided by the Border Air Quality Strategy.
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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<sub>2.5</sub> 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.