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Evaluation of particle resuspension in young children's breathing zone using stationary and robotic (PIPER) aerosol samplers

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Abstract

Development of asthma in young children may be associated with high exposure to particulate matter (PM). However, typical stationary samplers may not represent the personal exposure of children ages 3 and younger since they may not detect particles resuspended from the floor as children play, thus reducing our ability to correlate exposure and disease etiology. To address this, an autonomous robot, the Pretoddler Inhalable Particulate Environmental Robotic (PIPER) sampler, was developed to simulate the movements of children as they play on the floor. PIPER and a stationary sampler took simultaneous measurements of particle number concentration in six size channels using an optical particle counter and inhalable PM on filters in 65 homes in New Jersey, USA. To study particle resuspension, for each sampler we calculated the ratio of particle concentration measured while PIPER was moving to the average concentration of particles measured during a reference period when PIPER remained still.

For all investigated particle sizes, higher particle resuspension was observed by PIPER compared to the stationary sampler. In 71% of carpeted homes a more significant (at the $\alpha = 0.05$ level) resuspension of particles larger than 2.5 μm was observed by PIPER compared to the stationary sampler. Typically, particles larger than 2.5 μm were resuspended more efficiently than smaller particles, over both carpeted and bare floors. Additionally, in carpeted homes estimations of PM_{10}

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mass from the particle number concentrations measured on PIPER while it was moving were on average a factor of 1.54 higher compared to reference period when PIPER was not moving. For comparison, the stationary sampler measured an increase of PM_{2.5} mass by a factor of only 1.08 when PIPER was moving compared to a reference period. This demonstrates that PIPER is able to resuspend particles through movement, and provide a better characterization of the resuspended particles than stationary samplers. Accurate measurement of resuspended PM will improve estimates of children's total PM exposure.

Keywords

children's exposures to PM; robotic sampling platform; Pretoddler Inhalable Particulate Environmental Robotic sampler; PM size fractions; resuspension; floor type

1. Introduction

Exposure to airborne particulate matter (PM) has been associated with cardiovascular events (Miller *et al.*, 2007), lung inflammation (Raja *et al.*, 2010), and bronchitis as well as increased mortality in the case of PM₁₀ and PM_{2.5} (particulate matter with aerodynamic diameters smaller than 10 and 2.5 μm , respectively) (Samet *et al.*, 2000). PM can be produced indoors from activities such as cooking (Evans *et al.*, 2008), smoking (Jinot & Bayard, 1994), and burning candles (Weichenthal *et al.*, 2007) as well as by entering the residence from outdoors (Franck *et al.*, 2006). Also, resuspension of settled PM during regular household activities can result in PM levels as much as 17-fold greater than background levels (Ferro *et al.*, 2004). Asthma rates in children, have been found to be strongly correlated with the inhalation of particles < 1 μm in diameter (Franck *et al.*, 2011). Wong *et al.* found a relationship between indoor gas cooking and increase in asthma-like symptoms in preschool-aged children (Wong *et al.*, 2004). Increased outdoor PM levels have been associated with increased asthma-like symptoms among children 5 to 12 years old (Lewis *et al.*, 2013) and more frequent emergency room visits for breathing problems by inner-city children (Norris *et al.*, 1999). Our own recent research has observed an association between resuspended PM levels and asthma in children under 5 years of age (Ramagopal *et al.*, 2014).

These and other studies clearly indicate that children's exposure to PM could be detrimental to their health; however estimating exposure of young children to PM is a daunting task. Methods such as using a personal pump and filter samplers are not feasible for young children due to the weight and/or bulkiness of the equipment, and the potential choking hazard that they present because of young children's tendency to mouth objects. In addition, compliance with a study protocol could be problematic. Therefore, stationary samplers are often used to estimate exposures of children to PM. However, since people resuspend particles from the floor as they move around (Kubota & Higuchi, 2013; Mukai *et al.*, 2009; Qian *et al.*, 2008), stationary samplers are thought to underestimate this source of inhalable PM. This underrepresentation is likely to be especially dramatic in PM exposure estimates for young children because the recommended height of 110 centimeters for stationary samplers (EPA, 2003) is well above the breathing zone of children 3 years old and younger.

Additionally, since children often play on the floor this would cause resuspension of particles, and their actual exposures could be substantially higher than those measured by stationary samplers.

The Pretoddler Inhalable Particulate Environmental Robotic (PIPER) sampler was developed to improve our estimates of children's exposure to PM by acting as a surrogate when they play in their homes, especially on the floor; see (S. L. Shalat *et al.*, 2007) and (S. Shalat *et al.*, 2011) for a more detailed description. One of the benefits of PIPER is repeatability of its movement patterns over multiple sampling sessions and locations, thus removing the variability of individual children's movements on different days from repeated sampling and allowing for comparisons of particle resuspension under different sampling conditions. As part of our continuing effort to characterize PIPER and improve exposure estimates for young children, this study sought to investigate particle resuspension from flooring when PIPER is in motion by comparing PM size and concentration measured by equipment on PIPER with that measured simultaneously by identical equipment on the stationary sampler in the same room. Specifically, we sought to answer the following questions:

1. How does PIPER's motion affect the particle number concentrations and size distributions with respect to reference values measured by identical OPCs on PIPER and the stationary sampler when PIPER was motionless?
2. What is the effect of flooring type (carpet vs. bare floor) on the concentration and size distribution of particles measured?
3. How well do the PM_{2.5} and PM₁₀ mass estimates based on the particle number concentration agree between the two samplers (PIPER and stationary) when PIPER is in motion? How do they compare to the reference particle number concentration values in the same room when PIPER was motionless?
4. How well did the PM mass concentration estimated by OPCs correlate with filter-based inhalable particulate matter mass concentration?

2. Methods

2.1 Description of PIPER and stationary sampling setup

PIPER (Figure 1) is an autonomous robot designed to mimic the movements, including breathing zone height and speed, of young children as they play. Air sampling instruments such as samplers and pumps, optical particle counters (OPCs), and other devices can be placed on PIPER, with their inlets attached to a lift. During sampling, the lift can change in height from 20 to 84 cm above the floor to simulate changes in children's breathing zone heights that occur for different postures in children up to 3 years of age. PIPER moves autonomously on the floor while mimicking the average speed of motion of children 36 months old or younger. Six program files (ages 6 months to 1 year old, >1 year old to 2, and >2 years old up to 3 for each gender) were developed that can be run from a laptop and sent via wireless transmitter to direct PIPER's movements. Each file is designed to match the average fraction of time spent sitting, standing, and walking/running by girls and boys in a particular age group, as determined by analyzing videos of children of each age and gender

(S. Shalat, et al., 2011). Controlled experiments on clean flooring verified that PIPER itself, i.e., motors, wheels, etc., produce negligible amount of particulate matter. Previous PIPER studies have found that the concentration of inhalable PM measured by samplers on PIPER is higher by a factor of 2.30 in homes with carpeted floors and 1.12 in homes with bare floors than inhalable PM measured by a stationary sampler operated simultaneously (S. Shalat, et al., 2011); endotoxin levels measured by PIPER were nearly 3-fold higher than that those measured by the stationary sampler in carpeted homes (Wang *et al.*, 2012).

A full description of the sampling procedure for this study could be found in our earlier publication (S. Shalat, et al., 2011). Briefly, sampling took place in 65 homes in New Jersey, USA, from October 2009 to December 2010; each home had at least one child up to 36 months old. Measurements were performed, by both a stationary sampler and PIPER, in the room that the child played in most frequently; this room had carpet in 43 homes and bare floor in 22 homes. The stationary sampler and PIPER were each equipped with a TSI Aerotrak 8220 optical particle counter (OPC) which measured the number of particles per cubic meter in six size channels (0.3 to 0.5 μm , 0.5 to 1.0 μm , 1.0 to 2.5 μm , 2.5 to 5.0 μm , 5.0 to 10.0 μm , and greater than 10.0 μm) as one-minute average in each channel and operated at a sampling flow rate of 2.83 L/min. The inlet of the OPC on the stationary sampler was pointed upward, with conductive tubing attached, and the sampling point was fixed at a height of 110 cm above the floor. The OPC on PIPER was held at its base, while conductive sampling tubing extended to the lift where the sampling height varied from 20 cm to 84 cm during sampling. When PIPER was stationary, the sampling height was 20 cm above floor. Each sampler was also equipped with a Button inhalable aerosol sampler and Leland Legacy pump (SKC Inc., Eighty Four, PA). All optical particle counters were calibrated annually by the manufacturer (TSI Inc.) and the flow rates for all Leland Legacy sampling pumps was verified before and after each sampling period with a TSI Model 4199 flowmeter.

2.2 Measurement protocol

Graphic illustration of the sampling protocol with PIPER is shown in Figure 2. At the start of the measurements, both OPCs were turned on and allowed to run for 30 minutes with a 6-second delay between measurements, resulting in 26 one-minute particle number concentration averages. During those 30 minutes, PIPER remained still to allow most of the particles resuspended by the investigators to settle (Figure 2). The 30-minute period was chosen after preliminary tests with the OPC indicated that 30 minutes was enough time for particle number concentration to attain observed steady state concentrations. After 30 minutes, PIPER was turned on and it moved around the room for 30 more minutes, collecting 26 one-minute samples according to the age and gender profile of the child in a particular house while both OPCs recorded data. The OPCs were then shut off, but filter measurements were taken for 60 minutes on both samplers for additional comparisons (S. Shalat, et al., 2011). The pump connected to the Button sampler on the stationary sampler was started at the same point as PIPER's initial movements and ran for 60 minutes. Since PIPER can only hold two sampling instruments at once, and bioaerosol measurements were taken concurrently with the OPC measurements, filter measurements started 30 minutes later, at the end of the OPC measurement period as PIPER continued to run. The filters used

were 25 mm Teflon filters with 3.0 micron pore size. Sampling flow rate was 10.0 L/min, with a total filter sampling time of one hour.

2.3 Data analysis

For data analysis purposes, we defined samples 20-25 from the initial portion of the sampling period when PIPER was not moving (Figure 2) as the “reference period”. These six consecutive one-minute samples were chosen to represent the background particle because they are at the end of the still period, when most particles aerosolized by the investigators when the instruments were being set up would have settled down. Also, the selected time period represents approximately one quarter of the entire still period. We decided not to use the final sample of the still period, because particle number concentrations tended to increase due to the movements of the researchers as they prepared to turn PIPER on for the next portion of the sampling protocol. For data analysis, we compared particle number concentrations registered by the OPC on PIPER when it was moving with those when PIPER was still; this method allowed for comparison among homes with varying background particle number concentrations. Thus, for each 1-minute measurement period (t) while PIPER was moving and for each size channel (i) we calculated $RP_{PIPER}(ti)$, which is a ratio of particle number concentration measured by the OPC on PIPER vs. the average particle number concentration from the same OPC during the reference period:

$$RP_{PIPER}(ti) = \frac{\text{particle concentration measured by PIPER OPC } (ti)}{\text{average particle conc.during reference period by PIPER OPC } (i)} \quad (1)$$

We use this ratio rather than the absolute particle number concentration in order to estimate the effect of PIPER's movement on airborne particle number concentration measured by the OPC on PIPER.

An analogous ratio, RP_{STAT} , was defined and computed for each 1-minute measurement by the OPC on the stationary sampler:

$$RP_{STAT}(ti) = \frac{\text{particle concentration measured by stationary OPC } (ti)}{\text{average particle concentration during reference period by stationary OPC } (i)} \quad (2)$$

The use of separate ratios for PIPER and stationary sampler avoided any bias associated with different performance of the two OPCs. It should be noted that side-by-side tests of the OPCs indicated that the two instruments were highly correlated in their readings (r^2 values of 0.85 to 0.97 depending on channel) and precise (y-intercept values ranged from -0.2% to 10.1% of the highest value for each channel).

In addition, we also estimated PM mass concentrations for both stationary sampler and PIPER from the particle number concentration data. The total PM mass in each size channel was estimated by assuming that all particles within the channel were spheres with diameter equal to the middle value of the channel range and had a uniform density of 1.65 g/cm³ (Tittarelli *et al.*, 2008). The mass in channels 1-3 was added to estimate PM_{2.5} mass, while the mass in channels 1-5 was added to estimate PM₁₀ mass. Thus, for PIPER, the ratio of

$PM_{2.5}$ mass estimated for each minute of the sampling period to $PM_{2.5}$ mass estimated during the reference period, $RM_{2.5_{PIPER}}$ (mass ratio for $PM_{2.5}$) is defined as:

$$RM_{2.5_{PIPER}} = \frac{\text{estimated } PM_{2.5} \text{ mass concentration from OPC on PIPER}}{\text{av.estimated } PM_{2.5} \text{ mass concentration during ref.period from OPC on PIPER}} \quad (3)$$

Ratio $RM_{10_{PIPER}}$ for PM_{10} measurements with PIPER was defined in a similar manner.

Likewise, the ratio $RM_{2.5_{STAT}}$ was defined and computed to compare $PM_{2.5}$ estimated by the stationary OPC with the $PM_{2.5}$ estimated by the same OPC during the reference period:

$$RM_{2.5_{STAT}} = \frac{\text{estimated } PM_{2.5} \text{ mass concentration from stationary OPC}}{\text{av.estimated } PM_{2.5} \text{ mass concentration during ref.period stationary OPC}} \quad (4)$$

The ratio $RM_{10_{STAT}}$ for PM_{10} measurements by the stationary OPC was defined in the same way.

Due to equipment malfunctions, in nine homes OPC data could be downloaded from the instruments on PIPER but not from the stationary sampler; the data from these nine homes were included in calculations and plots involving PIPER. In summary, there were 56 homes with data for both the stationary sampler and PIPER and an additional nine homes where only PIPER data were available.

3. Results

3.1 Particle number concentration from each sampler

Figure 3 shows the particle number concentration ratios for each size channel, stratified by sampler and floor type. The three smaller size channels (0.3 to 0.5 μm , 0.5 to 1.0 μm , and 1.0 to 2.5 μm ; Figures 3a-c), and the three larger channels (Figures 3d-f) were grouped together respectively for comparison purposes.

For the three smaller size channels (particles between 0.3 and 2.5 μm ; Figures 3a-c) and bare floor, the average RP_{STAT} and RP_{PIPER} stayed constant and close to 1, or started close to 1 and increased toward the end of the sampling period. The average $RP_{STAT}(0.3-0.5 \mu\text{m})$ over bare floor is 0.936; $RP_{STAT}(0.5-1.0 \mu\text{m})$ and $RP_{STAT}(1.0-2.5 \mu\text{m})$ are similar at 0.948 and 0.985, respectively (Table 1).

However, RP_{STAT} decreased somewhat over the sampling period when sampling was performed over carpet. This trend is particularly pronounced for $RP_{STAT}(0.3-0.5 \mu\text{m})$ (Figure 3a): the ratio dropped from 0.94 to 0.81 during the 26 minute sampling period, with an average value of 0.856. $RP_{STAT}(0.5-1.0 \mu\text{m})$ and $RP_{STAT}(1.0-2.5 \mu\text{m})$ showed a similar decrease across the sampling period, with the value leveling off in the final 5 minutes. $RP_{PIPER}(0.3-0.5 \mu\text{m})$, $RP_{PIPER}(0.5-1.0 \mu\text{m})$, and $RP_{PIPER}(1.0-2.5 \mu\text{m})$ in carpeted homes show a different trend. In each case, the average RP_{PIPER} increased during the first 5 minutes of the sampling period, decreased for 2 minutes, increased in minute 8, then

generally dropped for the rest of the sampling period (the spike at minute 20 is discussed later).

As shown in Table 1, the average RP_{PIPER} for size channels 1-3 was always higher than the average RP_{STAT} for each floor type. RP_{PIPER} values were an average 5 to 7% higher each minute over bare floors than RP_{STAT} values and 11 to 14% higher over carpeted floors than the RP_{STAT} .

For the three larger size channels (particles greater than 2.5 μm ; Figures 3d-f), $RP_{STAT}(2.5-5.0 \mu\text{m})$ and $RP_{STAT}(5.0-10.0 \mu\text{m})$ over bare floor were fairly constant at 1.0 to 1.2 through most of the sampling period and increased over the last 8 minutes. $RP_{STAT}(>10.0 \mu\text{m})$ increased in minute 2, then decreased through the beginning of the period and levelled off in the second half. For PIPER samples over bare floors, $RP_{PIPER}(2.5-5.0 \mu\text{m})$ increased at the end of the period, while the overall picture was less clear for $RP_{PIPER}(5.0-10.0 \mu\text{m})$ and $RP_{PIPER}(>10.0 \mu\text{m})$.

Over carpet, $RP_{STAT}(2.5-5.0 \mu\text{m})$, $RP_{STAT}(5.0-10.0 \mu\text{m})$, and $RP_{STAT}(>10.0 \mu\text{m})$ showed a decrease in the first few minutes of the sampling period and leveling off at approximately 1.0, 1.2, and 1.4, respectively. Conversely, $RP_{PIPER}(2.5-5.0 \mu\text{m})$, $RP_{PIPER}(5.0-10.0 \mu\text{m})$, and $RP_{PIPER}(>10.0 \mu\text{m})$ increased during the first 5 minutes of the sampling period, and the values ranged from 1.3 to 1.6 ($RP_{PIPER}(2.5-5.0 \mu\text{m})$), 1.7 to 2.2 ($RP_{PIPER}(5.0-10.0 \mu\text{m})$), and 2.0 to 2.7 ($RP_{PIPER}(>10.0 \mu\text{m})$) throughout the sampling period.

A noticeable difference between the smaller and larger particle size channels is that RP is considerably greater for the larger particle size channels compared to the smaller size channels, especially as measured by PIPER on carpeted floor (Table 1). Most notably, $RP_{PIPER}(>10.0 \mu\text{m})$ over carpeted floors is 2.22; in other words, 2.22 times as many particles greater than 10 μm were measured, on average, by PIPER while PIPER was moving in carpeted rooms compared to the reference period, when PIPER was still. The stationary sampler measured an average of 1.52 times as many particles larger than 10 μm , while PIPER was moving, compared to the reference period. The difference in ratios for bare floors was less pronounced: average value of 1.38 for RP_{PIPER} and 1.25 for RP_{STAT} . For particles 5.0-10.0 μm in size and carpeted flooring, the difference in RP ratios obtained by the two devices can also be clearly seen in Figure 3e. Here, the average value for RP_{PIPER} was 1.88 compared with 1.20 for RP_{STAT} . In general, as seen in Figure 3 and Table 1, PIPER's average RP values for the three larger size channels were systematically higher than those of the stationary sampler: an average of 9 to 13% higher per minute over bare floor than the stationary sampler and an average of 35 to 57% higher over carpet.

The significance of the differences between the two samplers was analyzed using the Wilcoxon rank-sum test, where RP_{PIPER} and RP_{STAT} for each minute at each house were compared. Figure 4 shows the fraction of homes of each floor type for which the two samplers showed statistically significant ($p < 0.05$) differences in RP values. Little difference between the two floor types is seen for the smaller three channels, while the RP values were more often different in carpeted homes for the larger three channels. Furthermore, we took the average of each house's ($n=26$) RP_{STAT} and RP_{PIPER} values and

compared them across the two floor types with the Wilcoxon rank sum test. The stationary sampler's RP values were not significantly different between the two floor types for any of the six size channels. However, PIPER's RP values were significantly different for the largest two size channels (5.0 to $10\ \mu\text{m}$; $p=0.004$ and $> 10\ \mu\text{m}$; $p=0.001$).

3.2 Estimated PM mass from each sampler

The average estimated mass relative to that during the reference period for all homes for each minute of sampling are shown in Figure 5. Figure 5a shows estimated PM mass concentration ratios in rooms that had bare floor. $RM_{2.5_{STAT}}$ was slightly less than 1 for most of the sampling period and rises to just over 1 by the end of the 30 minutes (26 sample periods), with an average value of 0.959 ± 0.349 (Table 3). $RM_{2.5_{PIPER}}$ had an average value of 1.02 ± 0.359 , suggesting a slight change in the mass concentration of fine particles estimated by either sampler between the end of the reference period and the 30 minutes while PIPER was moving. $RM_{10_{STAT}}$, describing the estimated PM_{10} mass concentration compared to the reference mass period concentration, had an average value of 1.03 and $RM_{10_{PIPER}}$ had an average value of 1.13 (Table 3). The higher $RM_{10_{PIPER}}$ average was likely due to the sharp increases in the mass concentration ratio during minutes 18 and 24, which were driven by extremely high values in house #4. Overall, for bare floors all four ratios were rather similar in their minute-by-minute value and overall profile suggesting little resuspension of $PM_{2.5}$ and PM_{10} particles when PIPER was moving on bare floors. Nonetheless, the ratios measured by PIPER were consistently, even if only slightly, higher. The ratio of $RM_{2.5_{PIPER}}$ to $RM_{2.5_{STAT}}$ averaged over each minute was 1.06; in other words, PIPER measured an average of 6% more $PM_{2.5}$ mass per minute while moving compared to the reference period than the stationary sampler did. Likewise, for PM_{10} , PIPER measured on average 11% more PM_{10} mass per minute than the stationary sampler did compared to the reference period. With the data from house #4 removed, PIPER measures an average of 3% more PM_{10} mass per minute. This suggests that some particles, especially the larger ones that are resuspended by the motion of PIPER, were not registered by the stationary sampler, whereas they were observed by the OPC on PIPER. One can see a slight upward trend for all ratios during the last few minutes of measurement, likely caused by the researchers moving around to prepare for the next part of the experiment.

Figure 5b shows $RM_{2.5}$ and RM_{10} ratios for homes with carpeted flooring. The levels of $RM_{2.5_{STAT}}$, starting at 1 slowly decreased through most of the sampling period before leveling off in the last 5 minutes, with an average value of 0.914 ± 0.206 (Table 3). $RM_{10_{STAT}}$ started at around 1.2 and also initially decreased before leveling off at a value of around 1.1 for the last 9 minutes of the period, with an average overall value of 1.08 ± 0.332 . Meanwhile, $RM_{2.5_{PIPER}}$ showed a slight increase during the first 6 minutes of the sampling period, and then generally declined, with the exception of a spike at minute 20; it has an average value of 1.02 ± 0.359 . The value of $RM_{10_{PIPER}}$ over carpet was substantially higher than the other RM values over carpet for each measurement minute with an average value of 1.54 ± 1.41 . Overall, $RM_{10_{PIPER}}$ strongly increased in the first 5 minutes, when PIPER began to move, from 1.3 to 1.8, then fluctuated between 1.4 and 1.8 during the remaining sampling period.

As in homes with bare floor, compared to the reference period, the $PM_{2.5}$ and PM_{10} mass increase estimated by PIPER was higher than that from the stationary sampler. The difference was especially pronounced for PM_{10} particles: PIPER estimated an average of 12% higher increase in $PM_{2.5}$ compared to the stationary sampler, while the relative difference was as high as 43% for PM_{10} . The difference in the average $RM_{2.5_{STAT}}$ and $RM_{2.5_{PIPER}}$ between floor types was not significantly different ($p = 0.680$ for $RM_{2.5_{STAT}}$, $p = 0.623$ for $RM_{2.5_{PIPER}}$). However, RM_{10} was higher for homes with carpet than homes with bare floor for both the stationary sampler (1.08 vs. 1.03; $p = 0.197$) and PIPER (1.54 vs. 1.13; $p = 0.018$). In the latter case, the difference was statistically significant.

We carried out a statistical comparison of the RM values for the two samplers at each house as shown in Figure 6. For both $PM_{2.5}$ and PM_{10} , significant differences ($p < 0.05$) between the stationary sampler and PIPER were more frequently seen in the carpeted homes. The difference was most pronounced for PM_{10} , where there was a difference between the two samplers in 68% of carpeted homes but only 50% of bare floor homes.

3.3 Correlation of estimated PM mass with measured inhalable PM mass

Inhalable PM was collected on Teflon filters using Button samplers as described in the Methods section. We were curious to see whether the PM_{10} measurements by the OPCs could also serve as surrogates for inhalable particle measurements, once the PM_{10} data by the OPCs were compared to the inhalable particles mass collected at the respective stations: stationary and PIPER. The inhalable sampler on the stationary sampler was run for 60 minutes, beginning at the same time that PIPER started moving. The inhalable sampler on PIPER was also run for 60 minutes, but started at the conclusion of the OPC sampling period, and PIPER continued to run during the 60 minutes. Due to problems with the OPC on the stationary sampler at nine homes, we collected measurements on the stationary sampler at 18 homes with bare floor and 38 homes with carpet, while PIPER took measurements at 22 homes with bare floor and 43 homes with carpet.

PM_{10} mass estimates from the OPC data were plotted against the filter measurements of inhalable PM (not shown). In each case, the inhalable mass concentration measured by filters and PM_{10} mass estimated from OPC measurements exhibited a positive correlation ($r^2 = 0.27$ for the stationary sampler and $r^2 = 0.41$ for PIPER), and these correlations were statistically significant ($p < 0.05$) for both samplers. Slope coefficients were less than 1, indicating that the mass concentrations of inhalable PM by filter were higher than the PM_{10} as calculated from the OPC data. This certainly can be expected, given that the OPC PM_{10} data do not include particles larger than 10 μm .

4. Discussion

Although PIPER and the stationary sampler used identical sampling trains and sampled simultaneously, four factors could contribute to the observed differences in measurements. First, PIPER is a mobile sampling platform, sampling air as it moves across the floor. Although PIPER's weight and wheeled motion are different from a child's, the relative increase in particles observed when PIPER is in motion demonstrates that particles are resuspended by its motion. Second, PIPER has a variable inlet height. The maximum

sampling height on PIPER (84 cm) is less than the constant height of the stationary sampler (110 cm). Thus PIPER's sampling points are always closer to the floor. Third, PIPER's motion and variable inlet height enable it to sample multiple near-floor locations in a room, in contrast to the fixed location of the stationary sampler. Fourth, one limitation of PIPER is the coiling of the tubing on the lift. Therefore, a fraction of larger particles has been lost in the tubing during the experiment, and our estimates of particle concentration increase measured by PIPER may be underestimated, especially for $RM10_{PIPER}$, in contrast to the straight tubing on the stationary sampler. The differences in samplers result in significant differences in measured inhalable PM in the home.

Several homes in the data set had spikes in particle number concentration when PIPER was moving. The largest spikes were found in house #1 (carpet) for particles greater than 10 μm in diameter at minute 21, house #4 (bare floor) for particles larger than 1 μm at minutes 18 and 24, and house #14 (bare floor) for particles smaller than 2.5 μm in minutes 20 and 25. At these points in the data set the RP values reached 4 and above, with many values above 10 and the highest at 68. All of the spikes were seen on PIPER only, with no simultaneous spike in the stationary sampler data. The sources of these spikes are unknown but could have been caused by PIPER bumping into couches, plants, dust pockets on the floor, or other items in the room. It also suggests the possibility of short-term exposures to very high PM concentrations when a PM reservoir is perturbed by a child playing on the floor. The relationship of these high, transient exposures to children's health symptoms is unknown. Studies using stationary monitoring have found increases in PM concentrations (coarse and fine) have resulted in increased asthma symptoms (McCormack et al., 2009).

As shown in Table 1, average RP_{PIPER} values were systematically higher than RP_{STAT} values. This indicates that particles resuspended as PIPER moved were measured by the OPC on PIPER, but these particles were not detected as efficiently by the stationary sampler. The $RP_{STAT}(0.3-0.5 \mu\text{m})$, $RP_{STAT}(0.5-1.0 \mu\text{m})$, and $RP_{STAT}(1.0-2.5 \mu\text{m})$ values over both floor types were, on average, less than 1. However, the $RP_{STAT}(2.5-5.0 \mu\text{m})$, $RP_{STAT}(5.0-10.0 \mu\text{m})$, $RP_{STAT>(>10.0 \mu\text{m})$ values were larger than 1 for both floor types, with $RP_{STAT}(5.0-10.0 \mu\text{m})$ and $RP_{STAT>(>10.0 \mu\text{m})$ being substantially higher than 1, suggesting that the stationary sampler did detect the increase in larger particles caused by PIPER's movement, although the increase was not as high as that detected by PIPER. Table 2 also shows that the largest differences between RP values for the two floor types were seen by PIPER in the larger two size channels, confirming the preferential resuspension of larger particles (5.0 μm and larger) from carpet. The most striking result of the PM mass estimates from the OPC measurements is the very high value of $RM10_{PIPER}$ for carpeted flooring (Figure 5b). As noted, the average value of $RM10_{PIPER}$ throughout the sampling period was 1.54 (Table 3): considerably higher than any other combination of sampler, floor type, and PM mass size fraction. The fact that the average increase of PM_{10} was 54% as measured by the OPC on PIPER on carpet suggests that particles 2.5 to 10 μm in size are especially effectively resuspended by the movement of PIPER. This was also seen in the particle number concentration data and was the most pronounced for the largest particles (those greater than 10 μm ; Figure 2f). This observation is in agreement with other studies involving adults (e.g., (Cheng et al., 2010; Thatcher & Layton, 1995), despite PIPER's much lighter

weight. RM_{10} is always higher than $RM_{2.5}$ for each sampler, but the difference between the two is more pronounced over the carpet (Figure 5b), due to the propensity for larger particles to be resuspended more easily. Each sampler individually shows much more PM_{10} mass over carpeted floors than bare floors, suggesting that PIPER is especially resuspending particles $2.5\ \mu\text{m}$ and larger. However, $RM_{10\text{PIPER}}$ over bare floor was not nearly as high, suggesting that bare floor is not as effective a particle reservoir as the carpet is and that the particle resuspension rate from hard surfaces is much lower than that from carpeted floor. Reduced resuspension over bare floor compared with carpet is in line with previous studies (Mukai, et al., 2009; S. Shalat, et al., 2011); carpet is also associated with higher particle loads (Roberts *et al.*, 2004).

The average $RM_{2.5}$ value for the stationary sampler was less than 1, on both floor types, while RM_{10} was never less than 1 when averaged over the sampling period, for either sampler or either floor type. An RM value of less than 1 indicates that the mass of PM present during the reference period was higher than the mass present while PIPER was moving. Only the stationary sampler registered average $RM_{2.5}$ values of less than 1, suggesting that particles resuspended by PIPER's movement were not reaching the stationary sampler. At the same time, $RM_{2.5\text{PIPER}}$ was also only slightly above 1 for both floor types, implying that PIPER itself was not measuring much more $PM_{2.5}$ mass while moving compared to the reference period. The calculated p-values for the difference in $RM_{2.5}$ and RM_{10} by floor type listed in Section 3.2 also confirm that particles of $2.5\ \mu\text{m}$ and smaller are not efficiently resuspended from either type of floor.

Inhalable PM mass (measured with an inhalable sampler and filter) and mass estimated from particle number concentration measured by an OPC were compared to see if number concentration of particles $< 10\ \mu\text{m}$ can serve as a proxy for inhalable PM mass when filter measurements are not available or not practical. When measurements are pooled for both floor types, the r^2 value from the stationary sampler measurements is 0.27, which is slightly higher than the carpet values alone, but much lower than the r^2 for the bare floor values alone. On PIPER, the pooled PM_{10} r^2 value is 0.41, which is higher than both of the r^2 values for each floor type alone. The higher pooled value is likely driven by the carpet values, which have a much wider range of inhalable PM values from the filters, reinforcing the finding of higher amounts of resuspended particles over carpeted floors.

The comparison between the stationary sampler and PIPER may have been influenced by additional factors including the age and type of flooring, outdoor sources, and ventilation sources in the home. No measurement of air exchange rate or operation of heating/cooling systems was made in the homes, but the fixed location of the stationary sampler in contrast to PIPER's multiple location sampling may have contributed to some of the differences among homes that we observed. Additionally, carpets and bare floors are not uniform in their respective material characteristics, which influences resuspension rates (Goldasteh *et al.*, 2013; Mukai, et al., 2009): another fact that could have contributed to differences in particle resuspension among homes. We do not think that any changes in outdoor PM levels were substantial enough to affect our results, since sampling typically took place during mid-morning and early afternoon rather than during rush hour, our sampling times were relatively short, and most homes were not located on busy streets. Given the short time

period of sampling, we do not believe that variation in outdoor PM levels substantially affected our experiments; previous studies have shown that outdoor PM concentration is relatively stable mid-morning (Connell *et al.*, 2005; Moreno *et al.*, 2009). Furthermore, any changes in outdoor PM concentration that reached into the home would likely have been measured equally by both samplers. Although these factors were not investigated in the current study, PIPER may be used in future studies to identify the potential influence of these and other factors. Previous studies have used volunteers repeating activities such as prescribed walking on different carpet types to identify factors affecting resuspension (Rosati *et al.*, 2008). The reproducible motion and sampling programs executed by PIPER will introduce an additional level of control in similar experiments.

Additional environmental variables such as number of people residing in the home, type of heating system, and the presence of pets (cat or dog) were considered as possible influencing factors on particle resuspension. Based on the Wilcoxon rank sum test, *RP* and *RM* values were not significantly different between homes with and without pets, and between homes with three or fewer residents compared to those with four or more residents. A slight but significant difference in medians was seen in PIPER's measurements in homes heated with forced air versus those heated in other ways for particles of 2.5 to 5 μm and 5 to 10 μm ($p = 0.01$ to 0.03 , respectively), with homes heated by forced air having approximately 10% higher *RP* values. Likewise, the $RM10_{PIPER}$ was approximately 10% higher and significantly different ($p = 0.03$) in homes with forced air heat compared to those with other heating systems. Overall, we feel that although environmental variables likely affect absolute particle concentration, their effects are not clearly evident when normalizing particle number or mass concentrations to a reference period.

The use of a traditional stationary sampler may result in serious underestimation of the amount of PM in children's microenvironment, especially in homes with carpeted floors. When knowing PM exposures is important during the study of etiology of diseases, such as asthma, PIPER and similar technologies offer a method to obtain inhalation estimates that may be more accurate for young children compared to stationary sampling. This is strongly suggested in our recent publication on respiratory disorders, including asthma (Ramagopal, *et al.*, 2014). We also realize that PIPER is not a perfect surrogate for personal exposure estimates, due to possible differences in particle resuspension rates, resuspension mechanisms (wheels versus stepping, crawling, etc.), and changes in breathing zone height between PIPER and real children. An ongoing study by this research group is comparing PIPER's PM exposure estimates directly to PM exposure in 2-year-old children measured by a personal sampler.

5. Conclusions

This project demonstrated the differences between particle number concentration measurements taken on a stationary sampler and on a moving sampling robot, PIPER. PIPER's movement resulted in an increase in particle number concentration and the magnitude of that increase in each of six size channels was larger when measured by PIPER than by the stationary sampler. When PIPER was in motion, concentrations of larger particles ($>5.0 \mu\text{m}$) increased the most, where both samplers recorded number

concentrations 1 to 2.5 times higher than during the reference period. Accordingly, particle mass concentrations estimated from PIPER's OPC measurements were also higher than mass concentrations estimated from the stationary sampler. Since children typically play on the floor, we conclude that incorporating measurements of resuspended PM, known to be underestimated by stationary samplers, will provide a more representative estimate of their PM exposures. PIPER is a tool, developed to enable reproducible mobile sampling, to better characterize resuspended PM in children's homes.

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- A new mobile particulate matter sampler (PIPER) was compared to a stationary sampler.
- PIPER measured more resuspended particles compared to the stationary sampler.
- Simulated movement of children resulted in more resuspension of particles over carpet.
- PIPER estimated higher PM₁₀ concentrations compared to the stationary sampler.



Figure 1.
Pretoddler Inhalable Particulate Environmental Robotic (PIPER) sampler.

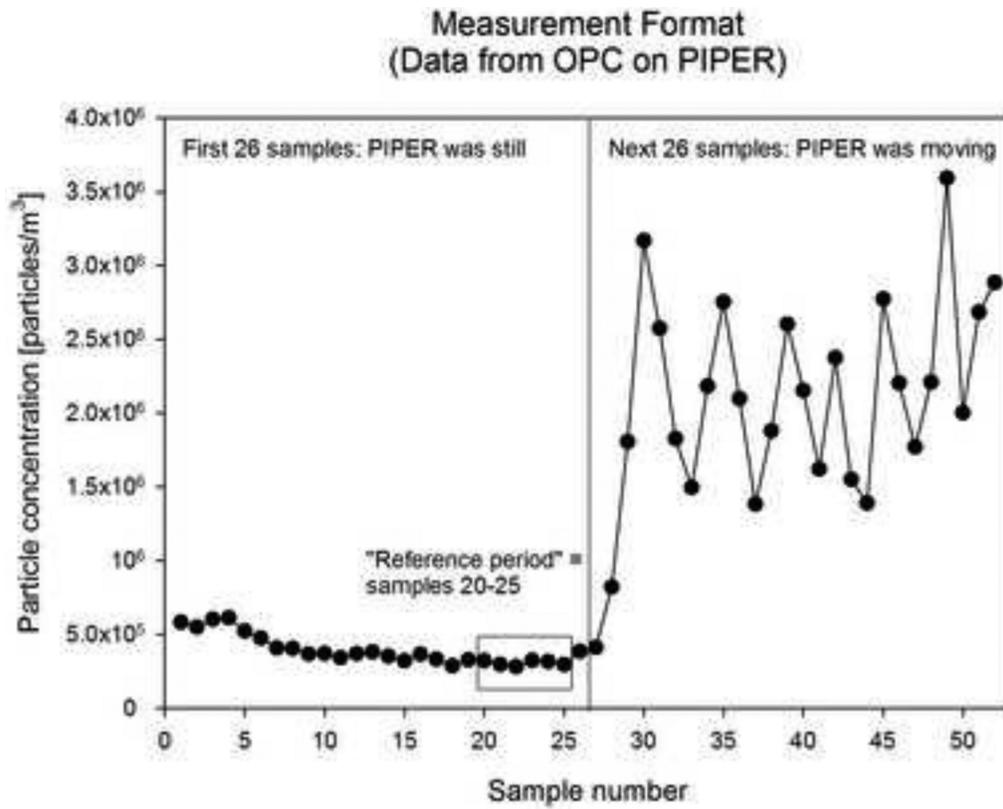


Figure 2.

Description of measurement setup. The sampling period started with 26 one-minute samples in which both OPCs were running, but PIPER was not moving. During the next 26 samples, PIPER moved about the room. The data shown are from the OPC sampler on PIPER, size channel 2.5 to 5.0 mm, at house number 36.

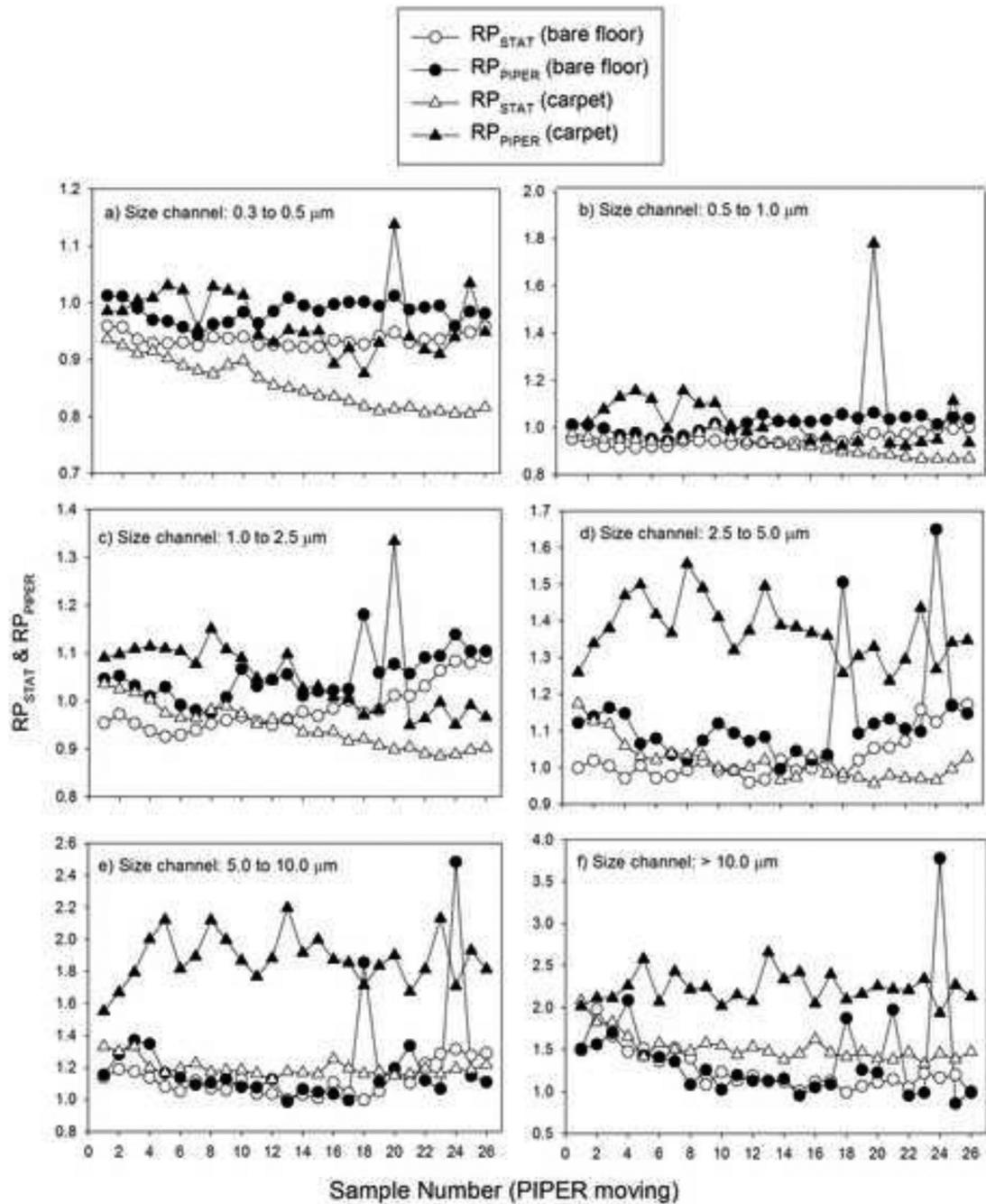


Figure 3. RP_{STAT} and RP_{PIPER} for each of the OPC's size channels. RP is the ratio of particle mass concentration while PIPER was moving to number concentration during the reference period (Equations 1 and 2). There were 43 homes with carpet and 22 homes with bare floor in the room that was sampled.

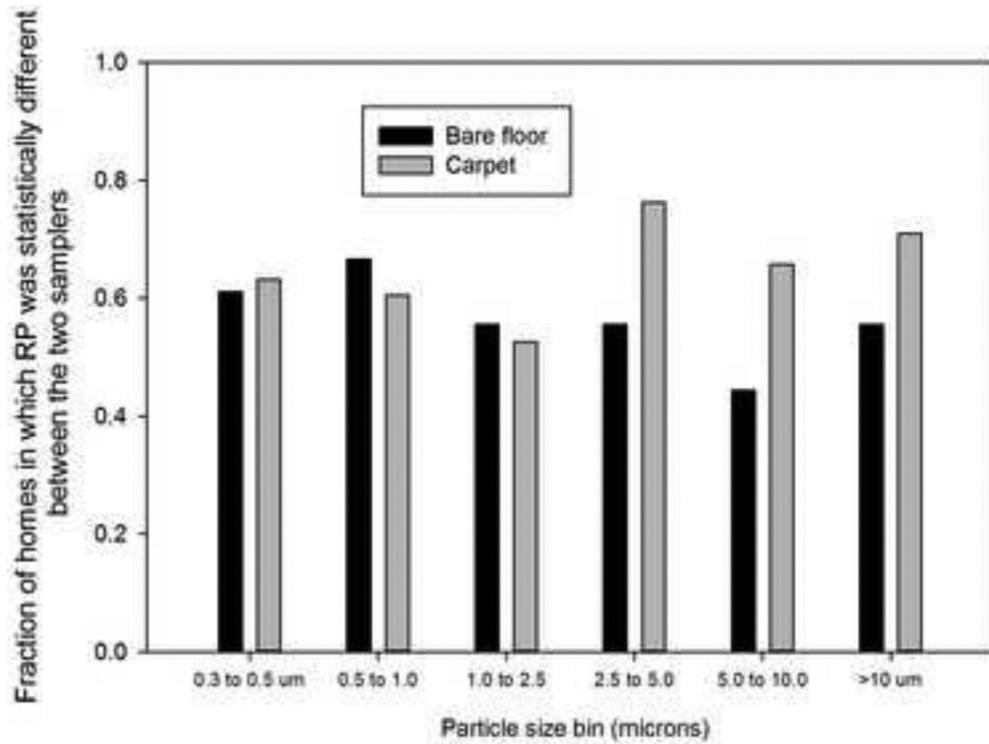


Figure 4. Comparison of the fraction of homes for which the average *RP* value was significantly different ($p < 0.05$) between the two samplers. *RP* is the ratio of particle mass concentration while PIPER was moving to number concentration during the reference period (Equations 1 and 2).

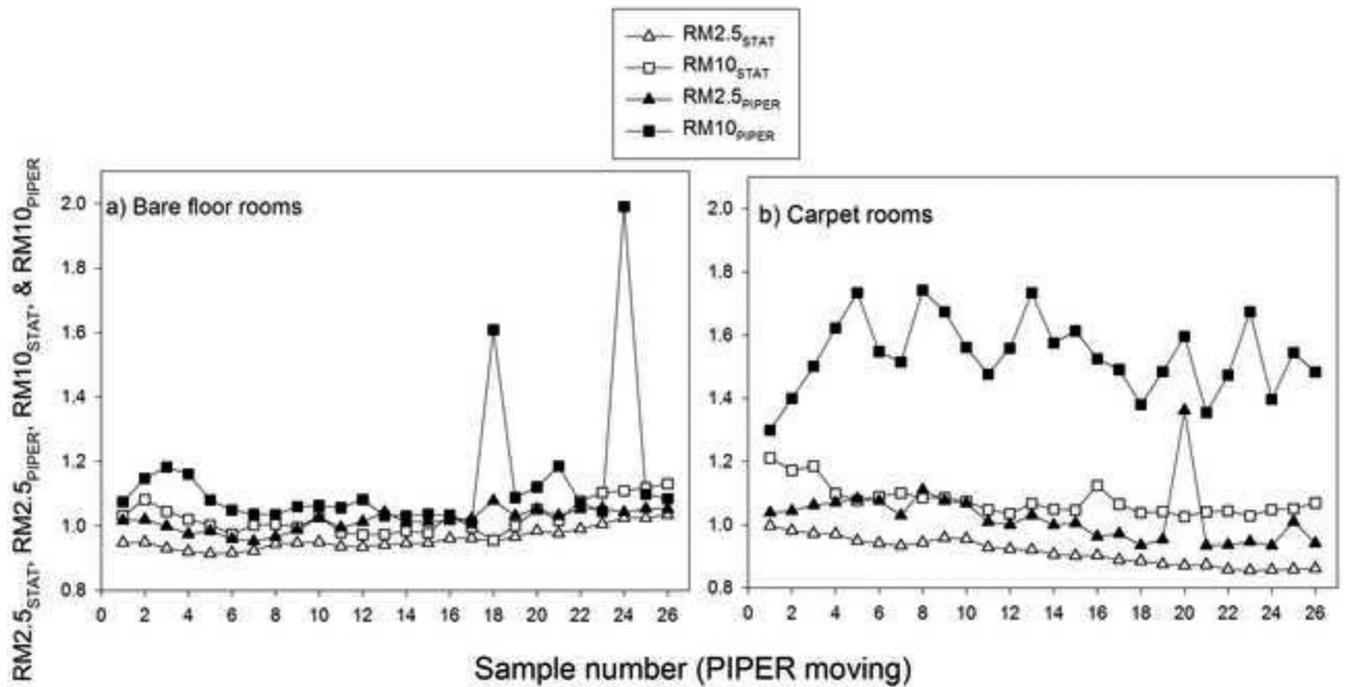


Figure 5.

$RM_{2.5}$ and RM_{10} measured by each sampler on bare floor and carpet. RM is the ratio of estimated mass while PIPER was moving to estimated mass during reference period (Equations 3 and 4). Out of 65 homes sampled, 43 had carpet and 22 had bare floor.

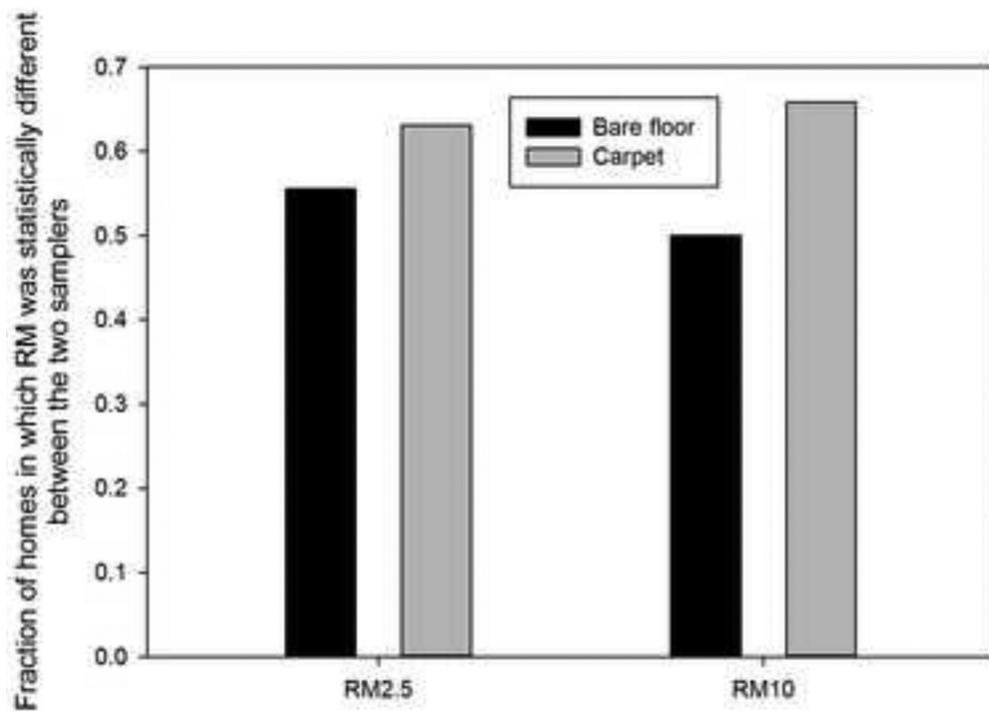


Figure 6. Comparison of the fraction of homes for which the average *RM* value was significantly different ($p < 0.05$) between the two samplers. *RM* is the ratio of estimated mass while PIPER was moving to estimated mass during reference period (Equations 3 and 4).

Table 1

Average RP (ratio of particle number concentration while PIPER moving to concentration during reference period; Equations 1 and 2) over all 26 samples. The standard deviation of RP was computed for each minute, size channel, sampler, and floor type across all homes.

Sampler	Floor type	0.3 to 0.5 μm	0.5 to 1.0 μm	1.0 to 2.5 μm	2.5 to 5.0 μm	5.0 to 10.0 μm	>10.0 μm
Stationary	Bare floor	0.936 ± 0.238	0.948 ± 0.340	0.985 ± 0.391	1.03 ± 0.495	1.12 ± 0.551	1.25 ± 0.651
	Carpet	0.856 ± 0.230	0.924 ± 0.283	0.947 ± 0.226	1.02 ± 0.301	1.20 ± 0.482	1.52 ± 0.783
PIPER	Bare floor	0.985 ± 0.231	1.02 ± 0.367	1.05 ± 0.422	1.13 ± 0.629	1.22 ± 0.814	1.38 ± 1.26
	Carpet	0.970 ± 0.404	1.05 ± 0.659	1.05 ± 0.463	1.37 ± 1.06	1.88 ± 1.93	2.22 ± 1.78

Table 2

P-values comparing the average *RP* values (ratio of particle number concentration while PIPER moving to concentration during reference period; Equations 1 and 2) for each size channel in carpeted homes versus average *RP* values in bare floor homes, for each sampler. P-values less than 0.05 are shown in bold.

	Stationary sampler	PIPER
0.3 to 0.5 μm	0.203	0.215
0.5 to 1.0 μm	1.000	0.683
1.0 to 2.5 μm	0.435	0.766
2.5 to 5.0 μm	0.330	0.103
5.0 to 10.0 μm	0.243	0.004
> 10.0 μm	0.081	0.001

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Table 3

Average *RM2.5* and *RM10* (ratio of estimated particle mass while PIPER was moving to estimated particle mass during reference period; Equations 3 and 4) for each sampler across the sampling period.

Sampler	Floor type	<i>RM2.5</i>	<i>RM10</i>
Stationary	Bare floor	0.959 ± 0.349	1.03 ± 0.420
	Carpet	0.914 ± 0.206	1.08 ± 0.332
PIPER	Bare floor	1.02 ± 0.359	1.13 ± 0.601
	Carpet	1.02 ± 0.384	1.54 ± 1.41

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