



A field application of a personal sensor for ultrafine particle exposure in children



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HIGHLIGHTS

- Children are able to wear a personal monitor for ultrafine particles (PUFP).
- The PUFP can measure personal exposure to UFP with spatial and temporal resolution.
- Children's exposure to UFP varies by microenvironment.

ARTICLE INFO

Article history:

Received 29 July 2014

Received in revised form 11 November 2014

Accepted 19 November 2014

Available online 10 December 2014

Editor: P. Kassomenos

Keywords:

Personal monitoring

Ultrafine particles

Children

ABSTRACT

Background: Ultrafine particles (UFPs) have been associated with adverse health outcomes in children, but studies are often limited by surrogate estimates of exposure. Accurately characterizing children's personal exposure to UFP is difficult due to the high spatiotemporal variability of UFP and children's time–activity patterns.

Objective: The objectives of this study were to conduct a field test of a personal sensor for UFP (PUFP) by measuring UFP exposure among children and assess the sensor's capabilities and limitations.

Methods: Children wore the sensor at school, during transit periods between school and home, and in their home for 2–4 h on 2 consecutive days and provided feedback regarding their experience with the sensor. The PUFP sensor recorded UFP number concentration at one second intervals and recorded GPS location allowing for comparisons of UFP exposure at homes, schools, and during transit. A mixed-effects linear model was used to compare the effect of microenvironment on personal UFP measurements.

Results: The overall total median personal exposure to UFP was 12,900 particles/cm³ (p/cm³). Median UFP exposure at homes, schools and during transit was 17,800, 11,900, and 13,600 p/cm³, respectively. Results of the mixed-effects model found that riding in a car and walking were significantly associated with 1.36 (95% CI 1.33–1.39) and 2.51 (95% CI 2.44–2.57) times higher UFP concentrations compared to the home.

Conclusions: The PUFP sensor can measure near real-time exposure to UFP with high spatiotemporal resolution. Children's exposure to UFP varies by location, with increased exposure during transit to and from school.

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1. Introduction

Despite advances in air pollution exposure assessment including the use of satellite data, sophisticated modeling approaches, and improvements in sensor technology, accurately measuring personal exposure remains a substantial challenge in studies of air pollution and human

health (Brauer, 2010). While the association between particulate matter (PM) and adverse respiratory health has been consistently demonstrated (Zanobetti et al., 2009; Millstein et al., 2004; McConnell et al., 2010; Gehring et al., 2010; Pope and Dockery, 2006; Dockery, 2009), the impact may be underestimated by surrogate measures of personal exposure which seldom considers all locations and activities which contribute to an individual's exposure (Van Roosbroeck et al., 2008; Meng et al., 2005). Most epidemiologic studies of PM use a combination of stationary monitoring and spatial models to estimate long-term exposure. Studies comparing these methods with personal exposure

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measurements have demonstrated that ambient concentrations and models for air pollutants with high spatiotemporal variability, including ultrafine particles (UFPs), often mischaracterize personal exposure (Nerriere et al., 2005; Samat et al., 2006; Diapouli et al., 2007). This discrepancy is particularly important for children, who are highly susceptible to these exposures due to their ongoing respiratory, cognitive, behavioral and neurologic development. Accurately assessing children's exposure to fine and ultrafine PM is intrinsically difficult due to the high spatiotemporal variability of UFP and the unique time–activity patterns of children, including time spent indoors at home and school, in vehicles, and walking, running, bicycling or playing near traffic sources during peak exposure periods (Brauer, 2010; Cattaneo et al., 2010). Therefore, children may experience high peak exposures over short time periods which cannot be captured by stationary monitoring (Liu et al., 2003; Hochstetler et al., 2011).

While studies of long-term exposure PM have consistently found deleterious cardiopulmonary health effects (Dockery, 2009; Pope and Dockery, 2006) fewer studies have characterized health effects associated with short-term exposure to air pollutants. More recent evidence is emerging, however, that short-term exposure to UFPs is associated with adverse respiratory health outcomes, especially among asthmatics. Short-term exposure (≤ 2 h) to UFPs emitted from diesel engines in real-world conditions has been linked to decreased lung function and increased inflammation among asthmatic adults (McCreanor et al., 2007). Another study of healthy adults demonstrated that short-term (2 h) exposure to air pollution during commuting activities was associated with decreased lung function and increased airway inflammation (Zuurbier et al., 2011). Healthy bicyclists were also found to have increased airway inflammation associated with PM number concentration (Strak et al., 2010).

Despite advances in technology allowing for the increased use of personal monitoring devices capable of assessing short-term PM exposure in near real-time, the method remains limited in its implementation due to the required time, labor, and costs associated with its use and the potential burden for study participants (Zou et al., 2009). Personal monitors have been utilized on a limited basis in studies of air pollutants, most often worn by adults (Janssen et al., 2005; Dons et al., 2012). In younger children, personal exposure to PM_{2.5} has been assessed using filter-based personal sampling with separate equipment for a pump, batteries, and filter or by using a nephelometer to assess real-time PM or black carbon concentrations (Delfino et al., 2006, 2004; Wallace et al., 2011). Using these tools, personal exposure to PM_{2.5} has been associated with increased airway inflammation and decreased FEV₁ among children with asthma (Delfino et al., 2006, 2004). UFP, whose contribution to PM_{2.5} mass is negligible, may have greater toxicity than larger particles due to their lung deposition, large particle number concentrations with increased surface area, and ability to translocate to other organs in the body (HEI Review Panel on Ultrafine Particles, 2013; Terzano et al., 2010). The impact of short-term personal exposure to UFPs on children's respiratory health, however, has not been well-studied due, in part, to a lack of personal monitoring devices, though recent studies have begun to characterize personal UFP exposure among children (Buonanno et al., 2013; Mazaheri et al., 2014).

In order to address these limitations a new, wearable, personal sensor for measuring exposure to UFP number concentration has been developed (www.enmont.com). This new device, henceforth referred to as a personal UFP (PUFP) sensor, is a UFP condensation particle counter (CPC) capable of measuring personal exposure to UFP number concentration (Son et al., 2013, 2011; He et al., 2013). The PUFP operates based on the principle of a CPC using water with comparable or better accuracy than the conventional, larger alcohol based CPC systems (Son et al., 2011; Hsiao et al., 2009; Choi and Son, 2009). In addition, the PUFP incorporates GPS technology to allow for accurate spatial characterization of exposure. The objective of this study was to conduct the first field test of this newly developed sensor with children in order to assess its capability to characterize personal UFP exposure. Further, the acceptability,

usability, and compliance of children and their caregivers to the PUFP were assessed.

2. Materials and methods

2.1. Personal ultrafine particle sensor

The PUFP (US patent # US 8,449,65) deployed in the field test is a condensation particle counter (CPC) with a total-system-volume of 1500 cm³, a weight of 1 kg, and approximately 6 h of continuous battery operation (for additional specifications see www.enmont.com). The PUFP is comprised of an evaporation–condensation-tube, a miniature diaphragm air pump, an optical detection module, a flow regulator, water tank, GPS, and battery pack in a plastic shell body (Fig. 1). The PUFP implements four electronic circuit boards to control sensor operation and data processing. The two central processing units on a board convert analog laser particle scattering signature to digital counting data along with the global positioning system (GPS).

The unit includes a lithium-polymer battery pack as a power source and an LCD monitor. An electronic circuit controls the battery and monitors its status to ensure the safety of the battery. In the case of an abnormal battery status including high-temperature, the electronic circuit automatically stops the sensor operation.

The PUFP has been tested at the Micro Thermofluidics Lab at the University of Cincinnati, the Particulate Matter Center at the University of Rochester (Rochester, NY), an industrial aerosol lab at MSP Corp (Shoreview, MN), and at the Underwriters Laboratory (Chicago, IL). Results of these tests have shown the PUFP to have a counting efficiency of 500,000 particles/cm³ with a lower size detection limit of 4.5 nm. In another study involving respirator leakage the PUFP produced comparable data to the TSI® Model 3007 CPC with a slope of ~ 1.16 and an R^2 of ~ 0.99 (He et al., 2013). Mobility tests, conducted under varying acceleration levels, demonstrated that the PUFP operates without noticeable performance degradation up to ± 4 –6-gravitational acceleration. UFP counts measured by the PUFP are well correlated with measurements obtained by a reference CPC (TSI Model 3022A) (data not shown). The inclusion of GPS technology allows for position and time data to be tagged to the measured UFP number concentration within 2 min and at one second intervals for excellent spatial and temporal resolutions.

2.2. Study population

Participants enrolled in the field test were recruited among children with asthma who attended one of three schools in Cincinnati, OH, most of whom had previously taken part in the Cincinnati Anti-Idling Campaign (CAIC) (Ryan et al., 2013; Eghbalnia et al., 2013). Schools participating in the CAIC study had either low or high bus traffic and were located either near or far from the closest major road. For the current study, nurses distributed information to former CAIC participants and also nonparticipants with asthma attending CAIC schools regarding the details of the field test, including its purpose, methods, and instructions on how to participate.

2.3. Field testing

Two personal sampling field test campaigns were conducted: one in fall 2012 and one in spring 2013. All study procedures and consent were approved by the University of Cincinnati Institutional Review Board (IRB). Two sensors were used during field testing; prior to their field deployment these sensors were calibrated side-by-side using a reference CPC (TSI3788) based on the procedure recommended by the National Institute of Standards and Technology. All deployed PUFP sensors had satisfied the calibration error requirement of less than 5% error over particle concentration ranges up to 500,000 p/cm³. In both campaigns, participants wore the PUFP sensor for 2–5 h on two consecutive days. Prior to each two-day field test, study personnel visited the homes of

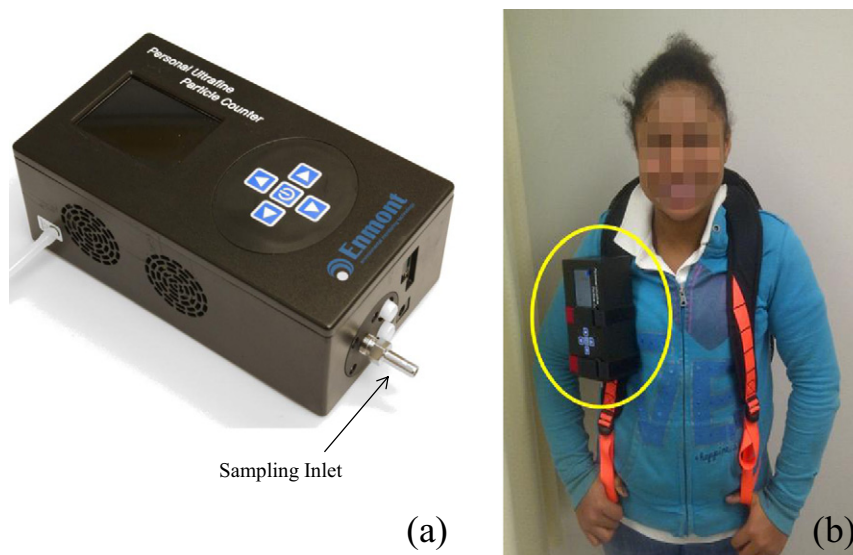


Fig. 1. PUFP (5th generation) sensor (a) attached to a backpack of field test participant (b).

participants to obtain written consent and to explain the study procedures. In addition, a baseline questionnaire was administered to parents to assess the participating child's respiratory symptoms, health care utilization, and home/environmental exposures including smoking by members of the household. Participants and their caregivers were also given a brief presentation regarding PUFP sensor operation and care. Field testing began prior to their departure from school and continued through the evening, after their arrival at home. This time period was chosen in order to assess short-term changes in ultrafine PM exposure at school, during transit periods between school and home, and in their home.

On each day of the field test, study personnel met each participant at school prior to dismissal to deliver the PUFP sensor. The PUFP sensor was fastened to the student's backpack near the breathing zone, and participants were asked to wear the sensor, or to be in close proximity to the sensor, at all times. The participant wore the PUFP sensor for the remainder of the day until evening. Simultaneous GPS and UFP concentration measurements were recorded at 1-second intervals, allowing for UFP exposure to be determined with high spatiotemporal resolution. After completing the field test, the parent and child were given a final survey regarding PUFP sensor acceptability and usability.

2.4. Personal UFP exposure by microenvironment

UFP concentration data and matching GPS coordinates were downloaded and projected in ArcGIS 10.1 software (ESRI). An exposure algorithm with spatial and temporal components was developed to categorize the personal UFP measurements into locations where children spent time during field testing. UFP measurements were categorized into four locations including 'school', 'home', 'transit', and 'other'. The address of each participant's home and school was geocoded and UFP concentrations at these locations were defined as follows: 1) 'school' – UFP measurement with corresponding GPS sampling coordinate <400 m of the participant's school, and 2) 'home' – UFP measurement with corresponding GPS coordinate <100 m of the participant's residence. Study questionnaires queried the participants regarding time spent at other locations and these were also geocoded. UFP concentrations recorded within <100 m of these locations were categorized as 'other' exposures (primarily after school care centers). UFP measurements recorded at GPS coordinates outside of the defined home, school, and "other" locations were assumed to occur during the participants'

transit between known stationary locations, primarily home and school, and were classified as 'transit' exposures.

At times during the sampling period, GPS coordinates were not recorded due to signal loss, chiefly a result of the sensor being indoors. In these circumstances, UFP measurements were recorded, but the subject's GPS coordinates and exposure category could not be determined based on the spatial component (GPS coordinates) alone. If GPS coordinates were not available during an interval of time between known microenvironment periods, those data points were categorized as within that exposure period. For example, GPS location may be recorded at school arrival, lost inside the school but regained once the participant sensor was worn outside. These data points without location information were classified as school exposure because the participant was known to be at school *before* and *after* the GPS signal was lost. Data points with missing GPS information that occurred between different microenvironments were categorized based on field observations by study personnel, questionnaire data (participant typical arrival time at home after school departure), time of known events (school dismissal), or the participant's activity pattern on the corresponding sampling day. Missing data between different microenvironments that could not be categorized based on these criteria were assigned to the previous known exposure period.

2.5. Statistical analysis and visualization of personal exposure

Though the PUFP sensor has previously been validated to measure UFP concentrations up to 500,000 p/cm³ with 95% accuracy in laboratory settings (constant room-temperature and humidity using electrically-neutralized UFP and hydrophilic UFP NaCl, unpublished data), in field conditions (variable temperature, humidity, and particle surface characteristics) measurement error is approximately ± 10% for concentrations exceeding 125,000 p/cm³. Therefore, recorded UFP concentrations exceeding 125,000 p/cm³ were replaced with 125,000 in the dataset prior to analyses.

Descriptive analyses were performed for the measured personal UFP exposure during the entirety of the personal monitoring (two days combined) and the distribution of personal UFP exposure for each microenvironment was examined. In order to examine the association between UFP exposure and each microenvironment and account for within-subject correlation, a mixed effects model was developed. In this model, the microenvironment of each subject for each sampled

time point was categorized into 6 possible locations: home, school, other, transit (bus), transit (car), transit (walk). These were coded as the only fixed effect in the model using 5 dummy variables, with home as the reference level. The random effects structure was such that each intercept was subject-specific. Prior to the analyses UFP concentrations were log-transformed to account for their skewed distribution. The coefficients of the mixed-effects linear model were back transformed and presented as the predicted fraction (95% confidence interval, CI) of UFP concentrations for each microenvironment compared to the home. In addition, to demonstrate the spatiotemporal UFP measurements obtained by the PUPF, personal UFP exposure for two selected subjects was visualized by plotting the UFP concentration and corresponding location using GIS software (ArcGIS 10.1, ESRI, Redlands, CA).

3. Results

3.1. Participant characteristics

The characteristics of the study participants and sampling times are summarized in Table 1. A total of 20 children participated in the field test; of these, 17 (85%) were African American and 14 (70%) were male. Participants' average age was 11.7 years and all had been diagnosed with asthma by a physician. Three modes of transportation to and from school were reported; 8 by bus, 6 by car and 6 walking. Of the 20 participants, 85% (n = 17) completed both days of field testing; the remainder (n = 3) completed one day of personal sampling. The overall average sampling duration and the average amount of sampling time at each microenvironment are presented in Table 1. Overall, the average total sampling time was 254 (± 87.9) min with time spent at home representing the longest sampling duration (117 ± 91.8 min). The average sampling times at school, transit, and other locations were 78.5 (± 53.5), 30.1 (± 28.7), and 31.0 (± 83.5) min, respectively (Table 1).

Overall, participating children spent, on average, 44% of the sampling period at home, 34% at school, 12% in transit, and 11% at other

locations. In general, the average contribution to participants' total UFP exposure during the field sampling was reflective of the time spent in each location and was highest for home (45%) and school (35%) and lowest for transit and other locations (14% and 7%, respectively). There were, however, some exceptions. For example, participant 20 spent only 12% of his sampling period at school but this time accounted for 42% of their total UFP. The contribution of UFP exposure from the school exceeded the time spent in schools for 12 participants whereas the time spent at home for 11 (55%) participants exceeded the contribution of home exposure to total UFP. The contribution of UFP exposure during transit exceeded the percentage of time spent in transit for 65% (n = 13) of study participants.

3.2. Personal exposure measurements

A summary of personal UFP exposure for all children is presented in Table 2. Less than 0.4% of all recorded UFP concentrations (1089/304,803) exceeded 125,000 p/cm³ and were replaced by 125,000 p/cm³ in subsequent analyses. Box-and-whisker plots (Fig. 2) present the distribution of the recorded UFP measurements on the log scale for all subjects and those measurements taken in each defined microenvironment. The distribution of the overall (combined) personal UFP measurements and those for each microenvironment was skewed due to high short-term exposure to UFP. The overall median exposure to UFP during personal sampling was 12,900 particles/cm³ (p/cm³). The overall median UFP exposure at schools (11,900 p/cm³) was less than that for both home (17,800 p/cm³) and transit (13,600 p/cm³) environments.

Estimated coefficients obtained from the mixed effects model are presented in Table 3 and were back transformed so that they could be interpreted as the multiplicative effect that each microenvironment has on total UFP exposure as compared to the home microenvironment. Results of the mixed model found significant differences in mean UFP concentrations between microenvironments within individuals. In particular, riding in a car and walking were estimated to result in 1.36 (95% CI 1.33–1.39) and 2.51 (95% CI 2.44–2.57) times higher UFP concentrations compared to the home. In contrast to these two microenvironments, taking the bus (0.77, 95% CI: 76–79) and being at school (0.90, 95% CI: 0.89–0.91) were associated with lower UFP exposure compared to home levels.

Spatiotemporal UFP concentration patterns for two selected participants were visualized by plotting their UFP concentrations by sampling time (Fig. 3) and location (Fig. 4). Both participants walked from school to their homes. Figs. 3a and 4a present data from participant 7 who attended school A and resides in an urban area near a major interstate highway (< 125 m). As shown in Fig. 3a, UFP concentrations exceed 100,000 p/cm³ at both school and home with concentrations during transit, in general, exceeding 50,000 p/cm³. Time spent indoors and outdoors at the home may account for the variability in UFP concentrations observed. Figs. 3b and 4b present data obtained for participant 17 who attended school B and resides in a residential neighborhood with few major roads. In general,

Table 1
Characteristics of field test participants.

Age [years, mean (range)]	11.7	(9.2–13.9)
Gender [#,%]		
Male	14	(70%)
Female	6	(30%)
Race [#,%]		
Black	17	(85%)
White	3	(15%)
ETS in home ^a	8	(40%)
Method of school transit		
Bus	8	(40%)
Car	6	(30%)
Walking	6	(30%)
Personal sampling duration [minutes, mean (SD)]		
School	78.5	(± 53.5)
Transit	30.1	(± 28.7)
Home	117	(± 91.8)
Others	31.0	(± 83.5)
Total	254.1	(± 87.9)
Primary heating method		
Gas furnace	14	(78%)
Electric furnace	4	(22%)
Central air conditioning		
Yes	7	(35%)
No	13	(65%)
Personal sampling testing days		
Two day field test	17	(85%)
One day field test	3	(15%)
School		
A	6	(30%)
B	8	(40%)
C	6	(30%)

^a Parental report of household member smoking in home (yes/no).

Table 2
Summary of UFP particle number concentration (p/cm³) by location.

Location	Mean (SD)	5th %-tile	25th %-tile	Median	75th %-tile	95th %-tile
Personal – overall	21,400 (25,100)	900	4900	12,900	26,000	80,200
School	19,800 (22,800)	1900	4900	11,900	24,300	74,600
Home	27,000 (28,300)	2800	6800	17,800	34,500	98,600
Others	4100 (5700)	600	800	1000	4700	17,500
Transit	21,400 (20,600)	3100	7500	13,600	28,200	71,600
Walking	38,100 (26,800)	9800	17,300	27,400	61,000	87,900
School bus	23,400 (20,000)	2300	9400	18,200	30,300	64,500
Car	11,700 (11,000)	3400	5700	8000	12,800	30,700

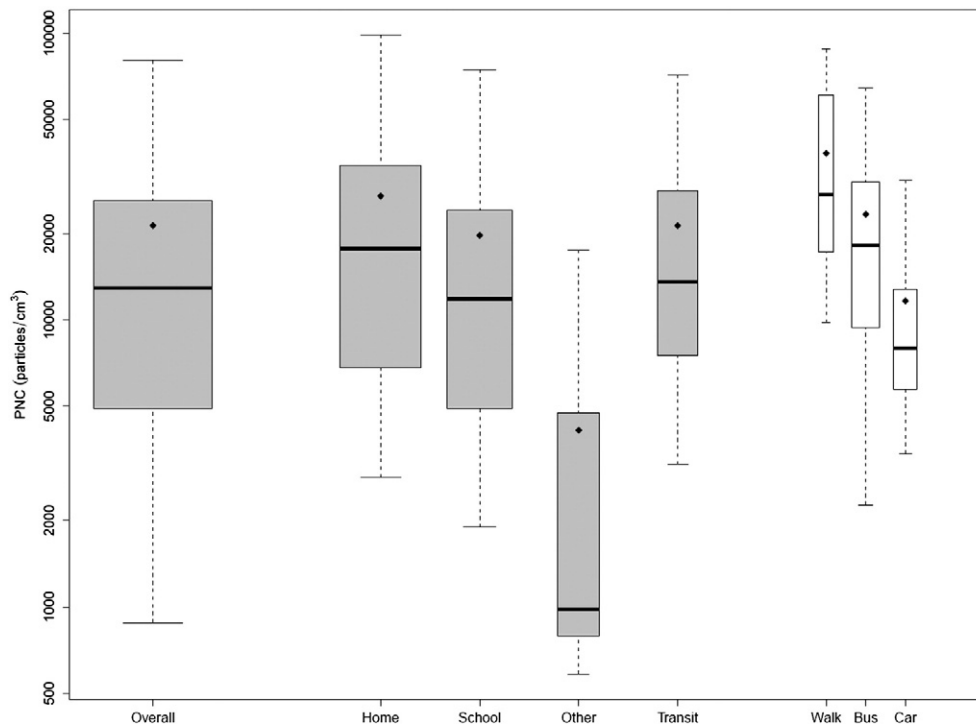


Fig. 2. Distribution of ultrafine particle number concentration by microenvironment. The width of each box plot is proportional to the square root of the sample size, diamonds represent the mean, solid lines indicate the median, boxes indicate the interquartile range, and whiskers indicate the 5th and 95th percentiles. Note that the y-axis is presented on the log scale.

personal UFP exposure was low ($<25,000$ p/cm³) at both school and home, but elevated ($25,000$ – $>100,000$ p/cm³) during transit. In addition, the transit route was located alongside multiple intersections where stop and go traffic patterns or idling vehicles may result in elevated UFP concentrations (Fig. 4b).

3.3. Participant evaluation

Participants provided feedback at the conclusion of the sampling period regarding their experience with the sensor. Overall, parents and children reported that the sensor was easy to use, with each group reporting a mean overall ease of use score of 4.4 out of 5. The most frequent comments regarding sensor limitations were related to noise with parents and children rating the sensor 2.1 and 2.2, respectively, out of 5), and weight (3.2 and 3.1, respectively). Despite these limitations, both parents and children reported the sensor's general ease of wear to be 3.8 and 3.6, respectively.

4. Discussion

The results of our field test demonstrate that the PUFF sensor is able to be worn by children in the context of an epidemiologic study while providing high-resolution spatiotemporal measurements of UFP. Further, results of the mixed linear model found significant individual differences in UFP concentrations in all microenvironments compared to

home concentrations. This suggests that inherent differences in subjects, in addition to microenvironments, contribute to UFP exposures.

An important component of our study was the PUFF sensor which was developed to characterize UFP number concentration exposure at 1-second resolution under conditions encountered in population-based studies. Commercially available particle counters are typically handheld devices used for area-based, rather than personal monitoring, and are limited by their size, weight, ruggedness, positional orientation, and particle size counting capabilities. In addition, conventional CPCs are limited by the liquid (alcohol or water) used as the fluid for condensation and can be easily flooded. As a result of these limitations, epidemiologic studies incorporating personal PM monitoring have frequently used filter-based collection of PM_{2.5} mass concentration. Filter-based PM_{2.5} monitoring, however, limits the ability to accurately characterize short-term and peak exposure to UFP, differentiate spatio-temporal gradients of UFP exposure, and distinguish the impact on human health of UFP from larger PM. More recently, personal monitors for surrogates of PM and UFP, including black carbon, have been developed to count PM indirectly using photometric technology. The photometric technique, however, has limited precision and accuracy due to the conversion of the total amount of scattered light from all PM to a particle concentration. Other studies have used nephelometers to measure PM exposure, but these also have limited precision and accuracy for measuring UFP as these instruments rely on light scattering to detect PM and are limited in their ability to detect PM smaller than the wavelength of their light source. UFPs, which are smaller than the wavelength of most personal nephelometers do not produce detectible scattering-light signal and therefore cannot be measured by these devices. Other devices using diffusion charging to measure UFP number concentration are also limited in their precision and accuracy. In contrast, the PUFF and other CPCs count individual scattered light produced by each particle.

Relatively few studies have conducted personal sampling of air pollutants in children. Using a nephelometer to assess personal exposure to PM_{2.5}, a study of 48 children with asthma in Windsor, CA found significant variability in PM_{2.5} exposures during a typical day (Van Ryswyk et al., 2014) with time sleeping associated with reduced PM_{2.5}

Table 3

Results of mixed effects linear model: estimated fraction of UFP exposure for each microenvironment compared to home.

Microenvironment	Estimated fraction (95% CI) of UFP exposure for each microenvironment compared to home
School	0.90 (0.89–0.91)
Other	0.30 (0.30–0.31)
Transit (walk)	2.51 (2.44–2.57)
Transit (bus)	0.77 (0.76–0.79)
Transit (car)	1.36 (1.33–1.39)

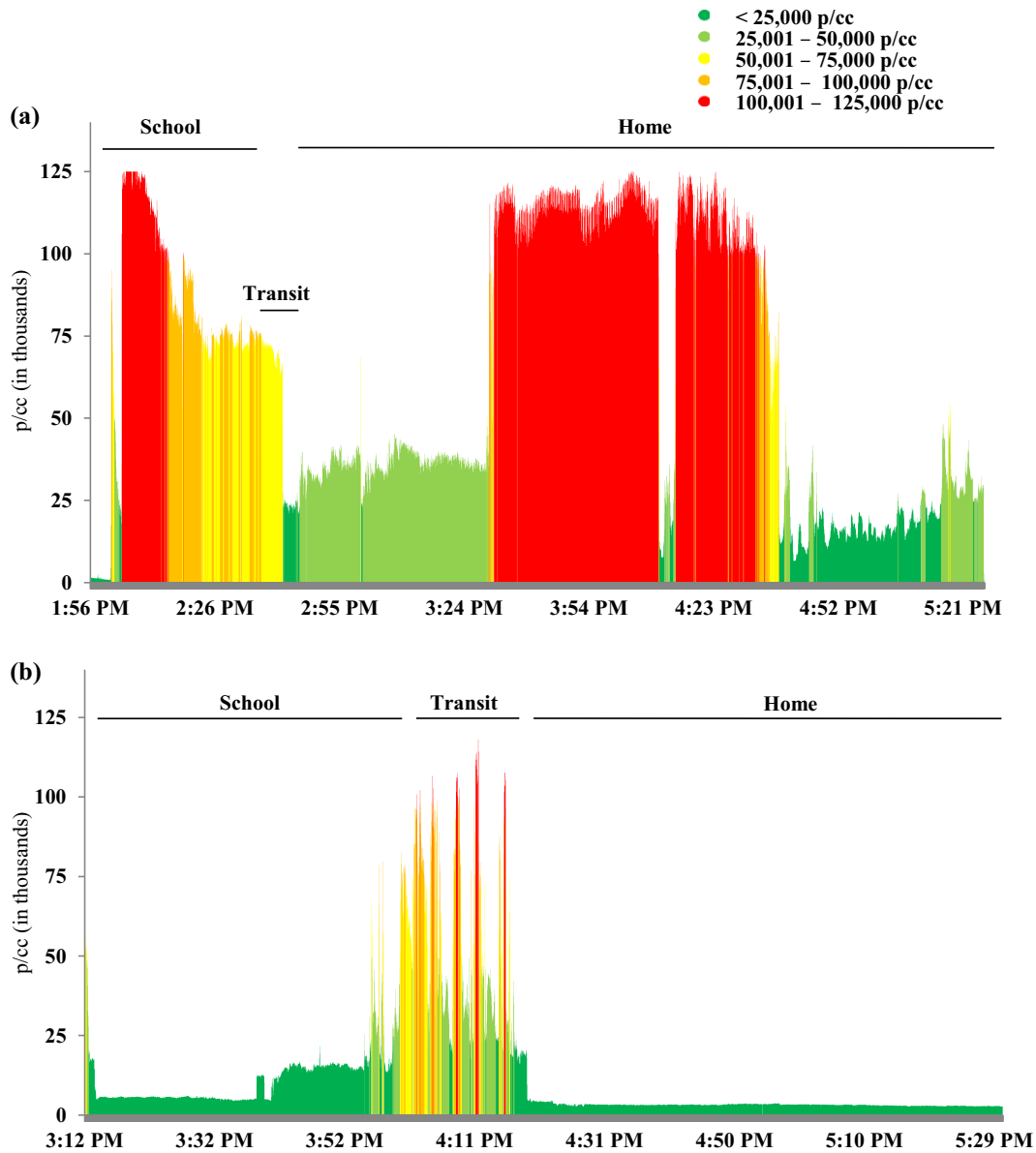


Fig. 3. Representative personal UFP exposure: participants 7 (a) and 17 (b) – personal UFP exposure by sampling time.

exposure and indoor playing, cleaning and food preparation associated with increased PM2.5 exposure. This same study also found that children spent approximately 70% of their time indoors at home which accounted for 52–66% of their PM2.5 exposure, depending upon season (Van Ryswyk et al., 2014). During our comparatively shorter sampling period, children spent 44% of their time at home, 34% at school, 12% in transit, and 11% in other locations and collectively the contribution of each microenvironment to overall UFP exposure was similar. A panel study of children with asthma in California also assessed personal exposure to PM2.5 using a nephelometer and found increased PM2.5 over 24 h to be associated with decreased lung function (Delfino et al., 2004).

We found that while the overall UFP exposure of children was similar across school, transit, and home environments (Fig. 2), there were significant differences in UFP concentrations by microenvironment when taking into account the within-subject correlation with a mixed effects model. The advantage of the mixed effects modeling approach is the ability to allow each child to have a unique and specific average UFP concentration (subject-specific intercept) while simultaneously estimating the effects of microenvironment for each subject. However, model interpretations must consider that the

coefficient estimates are specific to the subject and model. In our study, results of the mixed-effects model found that riding in a bus was associated with reduced UFP exposure compared to home concentrations, while riding in a car or walking were associated with increased UFP exposure relative to home concentrations. However, as shown in Fig. 2, the overall distribution of UFP exposure during bus riding and walking are similar, and UFP concentrations for both methods of transit are higher than while riding in a car. This apparent contradictory result is due to the subject-specific effects of children who ride in buses also having higher home UFP concentrations compared to the home concentrations of children who walk or ride in a car (data not shown). Hence, in our study, participants who rode a bus also have increased UFP concentration at their homes resulting in an estimated reduction in UFP exposure while riding the bus compared to their home concentrations. These findings lend further credence to the importance of individual time-activity patterns and specific spatial locations relative to UFP sources for an individual's exposure. The influence of time-activity patterns on personal exposure to UFP and black carbon (BC) has previously been studied in adults (Dons et al., 2012; Buonanno et al., 2014)

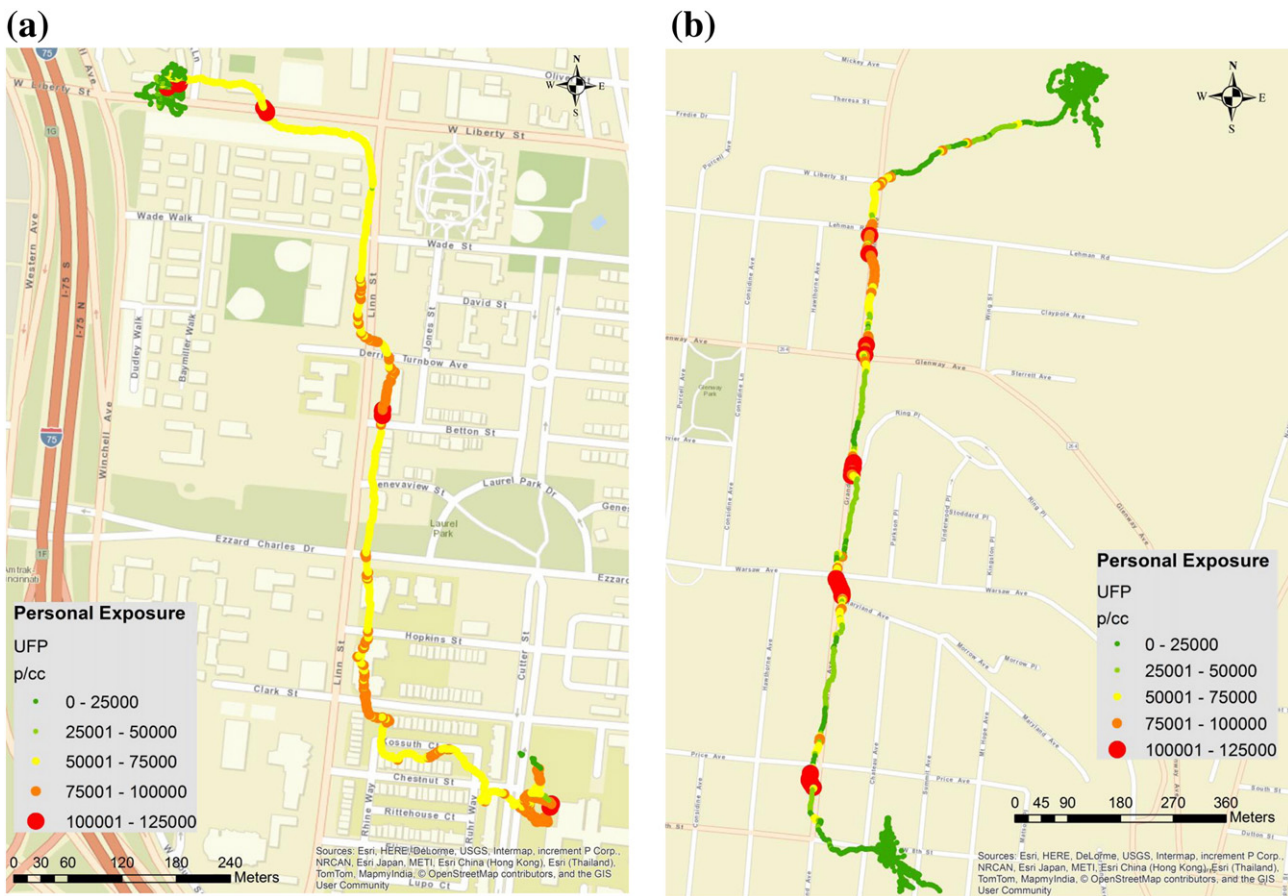


Fig. 4. Representative personal UFP exposure: participants 7 (a) and 17 (b) – sampling day 2. Personal UFP exposure by geospatial location.

and, indoor exposures to UFP were found to be higher in women due to indoor cooking whereas UFP exposure in men was greatest during transportation (Buonanno et al., 2014). Personal exposure to BC in adults was also shown to be highly dependent on time–activity patterns including time spent in transit and doing household activities (Dons et al., 2012). In 103 children ages 8–11, Buonanno et al. (2014) measured personal exposure to UFP using a commercially available diffusion charging UFP counter (NanoTracer, Philips) in Cassino, Italy and reported higher median and average UFP exposures than our study (22,000 p/cm³ and 58,000 p/cm³, respectively). Using self-reported time-activity data, spending time in transit and cooking/eating were identified as particularly important activities resulting in disproportionately high contributions to personal exposure relative to the overall time spent in these activities (Buonanno et al., 2014). Personal sampling for UFP exposure to children has also been conducted as part of the UPTECH project in Brisbane, Australia (Mazaheri et al., 2014). Using the Philips Aerasense NanoTracer device, average personal exposure to UFP at home, school and commuting (transit) was reported to be 10,500 p/cm³, 8530 p/cm³, and 13,700 p/cm³, respectively (Mazaheri et al., 2014). Our data, over a shorter time period, falls within the observed range of these studies, and in all three studies time spent in transit was a significant activity resulting in increased UFP exposure.

Similar to previous personal monitoring studies (Adams et al., 2009) we have defined microenvironments based on measurements taken in geographic proximity to the home, school, and other locations, with transit defined as neither of these. While we chose the proximity to

schools and homes (400 and 100 m, respectively) to reflect the potential size of each, it is possible that transit exposures may occur within these distances resulting in UFP measurements resulting from transit exposures to be assigned to the school and home microenvironments. Another limitation is the relatively short duration of our personal sampling period which encompassed school, transit, and home locations for just two partial days. Additional repeated sampling is likely required to more accurately characterize the contribution of each microenvironment to personal exposure and capture UFPs produced by cooking. The current PUFF sensor has a relatively short battery life (~6 h) and requires refilling the water reservoir after approximately 8 h of continuous use which is appropriate for characterizing acute exposure but limits its usefulness to measure UFP exposure over longer periods of time. Given our sample size, we were also unable to assess the contribution of specific indoor sources of UFP exposure, including the presence of smoking or gas stoves on personal UFP exposure. While GPS data allowed for spatiotemporal locations to be assessed, whether the child was indoors or outdoors was not known and missing GPS data due to loss of signal necessitated some assumptions regarding the location of the child relative to UFP measurements. Finally, user feedback indicated that the sensor was able to be worn by children, but participants indicated that its size, weight, and noise were drawbacks. Future planned research and development includes further modifications of the PUFF sensor to address these limitations and will facilitate its use in larger scale epidemiologic studies with a longer sampling duration.

In conclusion, in this field test of a new sensor for personal UFP exposure, we have demonstrated that children's personal exposure

to UFP varies by microenvironment. Our results reinforce the previously identified research need for methodology to capture exposure to UFPs with high spatial and temporal resolution. To date, epidemiologic studies of UFP exposure have been limited by potential exposure misclassification which may result in null health-related findings (HEI Review Panel on Ultrafine Particles, 2013). Furthermore, spatial models for UFP, including land-use regression, are limited in their ability to characterize indoor UFP exposure due to heating and cooking sources. The results of this study demonstrate that the PUF is able to measure, with high spatiotemporal variability, short-term and peak exposures to UFP allowing for additional research into the health effects of UFP exposure on children.

Author disclosures

The sensor technology development was funded by NIEHS grant U01ES16123 to the University of Cincinnati. This technology was subsequently licensed by the University of Cincinnati to EnMont, LLC which was founded and owned by Drs. Grace LeMasters, James Lockey and SangYoung Son.

Acknowledgements

The authors would like to thank the participants and their families. Funding was provided by the National Institute of Environmental Health Sciences R01ES020387.

References

- Adams C, Riggs P, Volckens J. Development of a method for personal, spatiotemporal exposure assessment. *J. Environ. Monit.* 2009;11:1331–9.
- Brauer M. How much, how long, what, and where: air pollution exposure assessment for epidemiologic studies of respiratory disease. *Proc. Am. Thorac. Soc.* 2010;7:111–5.
- Buonanno G, Stabile L, Morawska L, Russi A. Children exposure assessment to ultrafine particles and black carbon: the role of transport and cooking activities. *Atmos. Environ.* 2013;79:53–8.
- Buonanno G, Stabile L, Morawska L. Personal exposure to ultrafine particles: the influence of time–activity patterns. *Sci. Total Environ.* 2014;468–469:903–7.
- Cattaneo A, Taronna M, Garramone G, Peruzzo C, Schlitt C, Consonni D, et al. Comparison between personal and individual exposure to urban air pollutants. *Aerosol Sci. Technol.* 2010;44:370–9.
- Choi JY, Son SY, editors. Growth of ultrafine particles through a minichannel with capillary structure. *ICNMM Proceeding/CNMM2009-82248*; 2009. p. 841–7.
- Delfino RJ, Quintana PJ, Floro J, Gastanaga VM, Samimi BS, Kleinman MT, et al. Association of FEV1 in asthmatic children with personal and microenvironmental exposure to airborne particulate matter. *Environ. Health Perspect.* 2004;112:932–41.
- Delfino RJ, Staimer N, Gillen D, Tjoa T, Sioutas C, Fung K, et al. Personal and ambient air pollution is associated with increased exhaled nitric oxide in children with asthma. *Environ. Health Perspect.* 2006;114:1736–43.
- Diapoulis E, Chaloulakou A, Spyrellis N. Levels of ultrafine particles in different microenvironments—implications to children exposure. *Sci. Total Environ.* 2007;388:128–36.
- Dockery DW. Health effects of particulate air pollution. *Ann. Epidemiol.* 2009;19:257–63.
- Dons E, Int Panis L, Van Poppel M, Theunis J, Wets G. Personal exposure to Black Carbon in transport microenvironments. *Atmos. Environ.* 2012;55:392–8.
- Eghbalian C, Sharkey K, Garland-Porter D, Alam M, Crumpton M, Jones C, et al. A community-based participatory research partnership to reduce vehicle idling near public schools. *J. Environ. Health* 2013;75:14–9.
- Gehring U, Wijga AH, Brauer M, Fischer P, de Jongste JC, Kerkhof M, et al. Traffic-related air pollution and the development of asthma and allergies during the first 8 years of life. *Am. J. Respir. Crit. Care Med.* 2010;181:596–603.
- He X, Son S-Y, James K, Yermakov M, Reponen T, McKay RT, et al. Analytical performance issues: exploring a novel ultrafine particle counter for utilization in respiratory protection studies. *J. Occup. Environ. Hyg.* 2013;10:D52–4.
- HEI Review Panel on Ultrafine Particles. Understanding the health effects of ambient ultrafine particles. HEI Perspectives, 3Boston, MA.: Health Effects Institute; 2013.
- Hochstetler HA, Yermakov M, Reponen T, Ryan PH, Grinshpun SA. Aerosol particles generated by diesel-powered school buses at urban schools as a source of children's exposure. *Atmos. Environ.* 2011;45:1444–53.
- Hsiao T-C, Chen D-R, Son SY. Development of mini-cyclones as the size-selective inlet of miniature particle detectors. *J. Aerosol Sci.* 2009;40:481–91.
- Janssen NA, Lanki T, Hoek G, Vallius M, de Hartog JJ, Van Grieken R, et al. Associations between ambient, personal, and indoor exposure to fine particulate matter constituents in Dutch and Finnish panels of cardiovascular patients. *Occup. Environ. Med.* 2005;62:868–77.
- Liu LS, Box M, Kalman D, Kaufman J, Koenig J, Larson T, et al. Exposure assessment of particulate matter for susceptible populations in Seattle. *Environ. Health Perspect.* 2003;111:909–18.
- Mazaheri M, Clifford S, Jayaratne R, Mokhtar M, Fuoco F, Buonanno G, Morawska L. School children's personal exposure to ultrafine particles in the urban environment. *Environ. Sci. Technol.* 2014;48:113–20.
- McConnell R, Islam T, Shankardass K, Jerrett M, Lurmann F, Gilliland F, et al. Childhood incident asthma and traffic-related air pollution at home and school. *Environ. Health Perspect.* 2010;118:1021–6.
- McCreanor J, Cullinan P, Nieuwenhuijsen MJ, Stewart-Evans J, Malliarou E, Jarup L, et al. Respiratory effects of exposure to diesel traffic in persons with asthma. *N. Engl. J. Med.* 2007;357:2348–58.
- Meng QY, Turpin BJ, Polidori A, Lee JH, Weisel C, Morandi M, et al. PM2.5 of ambient origin: estimates and exposure errors relevant to PM epidemiology. *Environ. Sci. Technol.* 2005;39:5105–12.
- Millstein J, Gilliland F, Berhane K, Gauderman WJ, McConnell R, Avol E, et al. Effects of ambient air pollutants on asthma medication use and wheezing among fourth-grade school children from 12 Southern California communities enrolled in The Children's Health Study. *Arch. Environ. Health* 2004;59:505–14.
- Nerriere É, Zmirou-Navier D, Blanchard O, Momas I, Ladner J, Le Moulec Y, et al. Can we use fixed ambient air monitors to estimate population long-term exposure to air pollutants? The case of spatial variability in the Genotox ER study. *Environ. Res.* 2005;97:32–42.
- Pope III CA, Dockery DW. Health effects of fine particulate air pollution: lines that connect. *J. Air Waste Manag. Assoc.* 2006;56:709–42.
- Ryan P, Reponen T, Simmons M, Yermakov M, Sharkey K, Garland-Porter D, et al. The impact of an anti-idling campaign on outdoor air quality at four urban schools. *Environ. Sci. Process Impacts* 2013;15:2030–7.
- Sarnat SE, Coull BA, Schwartz J, Gold DR, Suh HH. Factors affecting the association between ambient concentrations and personal exposures to particles and gases. *Environ. Health Perspect.* 2006;114:649–54.
- Son SY, Lee JY, Fu H, Anand S, Romay F, Collins A. Personal and wearable ultrafine particle counter. Proceedings of the AAAR 30th Annual Conference, American Association for Aerosol Research Orlando, Florida, USA; 2011.
- Son S, Lee JY, Lockey J, LeMasters G., May 2013. Continuous droplet generator devices and methods. US Patent #: US8449657 B2.
- Strak M, Boogaard H, Meliefste K, Oldenwening M, Zuurbier M, Brunekreef B, et al. Respiratory health effects of ultrafine and fine particle exposure in cyclists. *Occup. Environ. Med.* 2010;67:118–24.
- Terzano C, Di Stefano F, Conti V, Graziani E, Petroianni A. Air pollution ultrafine particles: toxicity beyond the lung. *Eur. Rev. Med. Pharmacol. Sci.* 2010;14:809–21.
- Van Roosbroeck S, Li R, Hoek G, Lebrecht E, Brunekreef B, Spiegelman D. Traffic-related outdoor air pollution and respiratory symptoms in children: the impact of adjustment for exposure measurement error. *Epidemiology* 2008;19:409–16.
- Van Ryswyk K, Wheeler AJ, Wallace L, Kearney J, You H, Kulka R, et al. Impact of microenvironments and personal activities on personal PM2.5 exposures among asthmatic children. *J. Expo. Sci. Environ. Epidemiol.* 2014;24:260–8.
- Wallace LA, Wheeler AJ, Kearney J, Van Ryswyk K, You H, Kulka RH, et al. Validation of continuous particle monitors for personal, indoor, and outdoor exposures. *J. Expo. Sci. Environ. Epidemiol.* 2011;21:49–64.
- Zanobetti A, Franklin M, Koutrakis P, Schwartz J. Fine particulate air pollution and its components in association with cause-specific emergency admissions. *Environ. Health* 2009;8:58.
- Zou B, Wilson JG, Zhan FB, Zeng Y. Air pollution exposure assessment methods utilized in epidemiological studies. *J. Environ. Monit.* 2009;11:475–90.
- Zuurbier M, Hoek G, Oldenwening M, Meliefste K, van den Hazel P, Brunekreef B. Respiratory effects of commuters' exposure to air pollution in traffic. *Epidemiology* 2011;22:219–27.