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Measuring personal exposure to fine particulate matter (PM_{2.5}) among rural Honduran women: A field evaluation of the Ultrasonic Personal Aerosol Sampler (UPAS)



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ABSTRACT

We measured 24-hour average personal exposure to fine particulate matter (PM $_{2.5}$) among rural Honduran women using a lightweight, gravimetric monitor – the Ultrasonic Personal Aerosol Sampler (UPAS). Performance of the UPAS was compared with a commonly used gravimetric pump, cyclone, and filter sampling system. We observed strong agreement and correlation (Spearman $\rho=0.91$; PM $_{2.5}$ concentration range: $19-120\,\mu g/m^3$) between 43 paired measures, supporting the use of the UPAS as a personal exposure monitor for household air pollution studies.

1. Introduction

Exposure to household air pollution (HAP) from the combustion of solid fuels contributes annually to an estimated 2.6 (Health Effects Institute, 2018) to 3.8 (World Health Organization, 2018) million premature deaths primarily in low- and middle-income countries. Household air pollution is a mixture of many different airborne pollutants (Naeher et al., 2007), the most widely studied of which is PM_{2.5} (particulate matter with an aerodynamic diameter of 2.5 µm or less) (Balakrishnan et al., 2012). Conventionally, direct measurements of personal exposure to PM have involved devices that are cumbersome to wear, have limited battery life, and are therefore not well-adapted for use in resource-constrained and often remote developing country contexts. As such, many household air pollution-related studies rely on alternative approaches to estimate personal exposure to PM2.5. For example, proxy measures of exposure (Yu et al., 2018; Siddharthan et al., 2018; Amaral et al., 2017) - like self-reported stove and/or fuel type or kitchen area measurements of PM2.5 - are often used in place of wearable PM samplers and are prone to exposure misclassification (Clark et al., 2013; HEI Household Air Pollution Working Group, 2018). Further, without collection of PM mass, it is difficult to quantify the

contributions of multiple PM sources to individual exposure and to attribute exposure reductions to specific policies or interventions (Lai et al., 2018).

Instrumentation for PM_{2.5} gravimetric personal exposure measurement was originally developed for occupational settings (Volckens et al., 2017). Common practice involves collecting integrated personal PM_{2.5} samples using sampling pumps powered by batteries that are worn at the hip or in a backpack and connected with tubing to a size-selective device placed near the breathing zone of participants (Balakrishnan et al., 2012; Clark et al., 2013; Lai et al., 2018). These samplers are typically heavy and bulky. Sampling periods typically range from 8 to 48 h (samplers are removed and placed in proximity to the subject when bathing, sleeping, breast feeding, etc.) (Balakrishnan et al., 2012). Innovations in personal PM exposure assessment in recent years, including smaller and quieter monitors with longer battery life, overcome multiple barriers to more reliable, longer-lasting, and more convenient personal sampling.

In this paper, we describe a field-based evaluation of the Ultrasonic Personal Aerosol Sampler (UPAS, Access Sensor Technologies, Fort Collins, USA), one such next-generation personal PM monitor, among female study participants in rural Honduras who use wood-burning

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cookstoves. The UPAS includes a miniature piezoelectric pump to draw air through the device, a mass flow controller to regulate sampling rates, and a cyclone customized to capture an integrated PM2.5 sample on filter media housed within the device (Volckens et al., 2017). The UPAS weighs approximately 0.2 kg, is the size of a large cell phone $(9.7 \times 5.1 \times 2.5 \,\mathrm{cm})$, and is relatively quiet (< 40 dB at 20 cm) (Volckens et al., 2017). We compare the performance of this monitor to a more common setup: a personal sampling pump coupled via tubing to a cyclone and filter cassette. Specifically, we compare the PM_{2.5} concentration resulting from gravimetric analysis of filters used to collect PM_{2.5} mass. The UPAS has been evaluated, and has performed well, when compared to standard methods in both the laboratory (Volckens et al., 2017) and in a field setting with stationary measurements (Arku et al., 2018); however, the UPAS has not been evaluated against accepted technologies for personal measurements of PM2.5 mass in a field setting. Our objective was to evaluate the UPAS as a personal PM2.5 exposure monitor in rural environments where household air pollution is a concern by comparing it with conventional personal samplers.

2. Methods

This study was approved by the Colorado State University Institutional Review Board (#12-3870H); verbal informed consent was obtained from all participants.

2.1. Study location

Data were collected from April 3 to May 4, 2017 in rural Honduras. We evaluated personal exposure to PM_{2.5} mass among a convenience sample of 49 women participating in a cookstove intervention trial in communities near the town of La Esperanza, Department of Intibucá, located in western Honduras. Briefly, the larger randomized controlled trial (PI: Clark, NIH ES022269) in which this evaluation is nested seeks to characterize use and barriers to use of a cookstove intervention and to evaluate the impacts of the intervention on household air pollution exposure and cardiovascular health. Both the traditional and the intervention (*Justa*) stoves evaluated in the trial were wood-burning.

2.2. Personal exposure assessment

Simultaneous gravimetric samples were collected using one of four UPAS units and a common setup consisting of a pump connected by tubing to a cyclone affixed to a cassette containing a filter. Samples were collected on primary cooks for 24h during routine household activities (Fig. 1). Both samplers collected PM2.5 mass on polytetrafluoroethylene (PTFE)-coated, 37 mm, borosilicate glass fiber film filters (Pall Inc., Ann Arbor, MI, USA). The standard gravimetric setup consisted of a SCC 1.062 Triplex Cyclone (Mesa Labs, Lakewood, CO, USA) with attached filter cassette connected to an AirChek XR5000 (SKC Inc., Eighty Four, PA, USA) pump calibrated to operate at 1.5 L per minute, heretofore referred to as the "pump and filter." Flow rates for the pump and filter system were pre-calibrated and checked post-deployment in the field using a DryCal DC-Lite (Bios International Corporations, Butler NJ, USA) primary flow calibrator. The UPAS flow rates, which are logged internally, were calibrated to 1 L per minute before deployment and evaluated at the end of the sampling campaign by the manufacturer using a primary flow standard (F101D, Bronkhorst, Netherlands) and validated using a second primary standard (Alicat, USA). For both the pump and filter system and the UPAS, preto post-sampling flow rates did not deviate by > 10%.

The AirChek XR5000 sampling pump was placed in a small bag worn by the participant and connected via tubing to the cyclone; both the cyclone (for the pump and filter) and UPAS were attached near the breathing zone of the participant (Fig. 1) with sampling inlets at the same height and placed as close together as possible. Participants were asked to continue their normal daily routine while wearing the



Fig. 1. A participant wearing both the UPAS and the typical pump and filter gravimetric samplers. Note that both the cyclone from the pump and filter setup and the UPAS are approximately located near the participant's breathing zone.

instruments and to remove and place the instruments nearby while bathing or sleeping. Participants self-reported during the post-sampling interview the duration (in minutes) and frequency of exposure equipment removal, other than to bathe or sleep. Eight field blanks were collected throughout the study for each sampler type. After sampling, all filters were kept in a freezer in Honduras and at Colorado State University, where the filters were transported for weighing.

2.3. Gravimetric analyses

Filters from both sampling systems were removed from cold storage and placed in an equilibration chamber for at least 24 h prior to weighing on a Mettler Toledo MX5 Microbalance (Mettler Toledo, Columbus, Ohio) with 1 μg resolution. Prior to each measurement, static was discharged using a Mettler Toledo Antistatic U Ionizer for 10 s. Weights were taken in duplicate. If the weights differed by $>5~\mu g$, the filter was weighed a third time and the average of all three values was used. All weighing occurred at Colorado State University, Fort Collins, USA. $PM_{2.5}$ concentrations were estimated by dividing the blank-corrected filter mass by the volume of air sampled over the measurement period. The limit of detection (LOD, in grams) for each sampler was estimated by adding the mean mass of the field blanks to three times the standard deviation of field blank masses (United States Environmental Protection Agency, 2016).

2.4. Statistical analyses

Descriptive statistics for 24-hour $PM_{2.5}$ concentrations were calculated for both samplers. We also calculated Spearman correlation coefficients and created Bland-Altman plots (Altman and Bland, 1983) to estimate correlation and agreement between measurements collected by the two types of samplers. Finally, we calculated the root mean squared error (RMSE) and bias of measurements made by the UPAS. Bias was calculated as the mean difference of the paired pump and filter and UPAS $PM_{2.5}$ concentrations. All analyses were conducted in R (version 3.4.2, the R Foundation for Statistical Computing).

3. Results

Of the 49 paired samples attempted, 43 were successfully obtained. Four UPAS samples were not included for analysis because local power outages resulted in failed overnight charging and, thus, short sample

Table 1 Comparison of $PM_{2.5}$ personal exposures ($\mu g/m^3$) measured by SCC 1.062 Triplex Cyclones with SKC AirChek XR5000 pumps ("Pump and Filter") and the Access Sensor Technologies UPAS.

	Mean	SD	Median	IQR	Min	Max	N
Pump and filter	60.2	25.7	51.8	32.5	23.2	131.1	43
UPAS	52.5	19.9	45.7	30.0	19.0	96.8	

durations (12.6–15.1 h). Two pump and filter samples were discarded; one lost mass after sampling, indicating either a sampling or handling error, while the second had signs of oversampling and particle deposition patterns on the filter unlike all other filters (i.e., large particles or clumping of particles were visible with uneven deposition). These six paired samples were excluded from further analysis. The average mass of UPAS and pump and filter blanks was 15 μg (SD = 4, n = 8) and 11 μg (SD = 10, n = 8), respectively; estimated LODs were 27 μg for the UPAS and 41 μg for the pump and filter samplers. All sample filter masses were above the LOD for the respective devices.

Twenty-one participants (43% of those wearing both instruments) reported removing the equipment for $1{\text -}2\,h$ (4 to 8% of the total sampling time) on average. The concentration difference between measurements made by the UPAS and the pump and filter samplers among those participants who did and did not report removing samplers was not significantly different (Wilcoxon rank sum test, p=0.17).

Descriptive statistics by sampler type are provided in Table 1. Arithmetic mean $PM_{2.5}$ concentrations measured by the UPAS were, on average, slightly lower than those of the pump and filter (52.5 \pm 19.9 vs 60.2 \pm 25.7 µg/m³, respectively).

Mass concentrations estimated from UPAS samples were highly correlated with those from the pump and filter (Fig. 2, Spearman $\rho=0.91,\ 95\%$ CI: 0.84, 0.95). A Bland-Altman plot, which evaluates agreement between two types of measurements, found only one point outside of the limits of agreement (Fig. 3). When compared to the pump and filter, the RMSE of the UPAS measurements was approximately $13.3\,\mu\text{g/m}^3$; the bias was $7.7\,\mu\text{g/m}^3$.

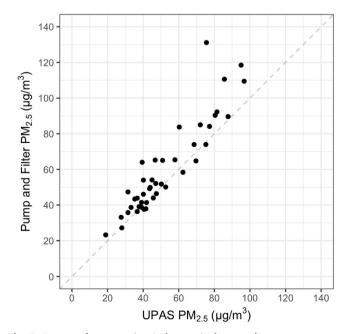


Fig. 2. A scatterplot comparing 24-hour paired personal exposure measurements collected with the UPAS (x-axis) and the Pump and Filter (y-axis) among 43 Honduran female cooks (Spearman $\rho=0.91,\,95\%$ CI: 0.84, 0.95). The dashed line is a 1:1 line.

4. Discussion

Our findings are consistent with previous laboratory and field-based evaluations of the UPAS. During the development of the UPAS, its performance was compared against that of the SKC Personal Exposure Monitor (PEM) and a federal equivalence method (FEM, URG Cyclone) in a laboratory chamber. Laboratory testing indicated highly linear relationships between the UPAS and the FEM ($r^2=0.996$), similar to the relationship of the PEM and FEM ($r^2=0.998$) (Volckens et al., 2017). These laboratory-based relationships are stronger than our field-based results, likely due to homogenous aerosol and highly controlled conditions typical of laboratory evaluations.

The strength of the correlation between personal exposure concentrations measured using the UPAS and the pump and filter in our study was similar to results from a recent field evaluation of stationary UPAS measurements compared with Harvard Impactors (Pearson's r = 0.91, 95% CI: 0.84, 0.95). In that field evaluation, which was conducted as preliminary work for the ten country Prospective Urban Rural Epidemiology (PURE) study, researchers collocated samplers in 43 rural Indian kitchens for either 24 or 48 h (Arku et al., 2018). Concentrations measured by PURE ranged up to 350 µg/m³, which was higher than what we observed in Honduras (max: 131 µg/m³), and likely due to typically higher concentrations measured in kitchens compared to measuring personal exposure (most cooks do not spend the entire cooking period or day in the kitchen). In the PURE study, the UPAS slightly overestimated concentrations when compared to the Harvard Impactor, contrary to what we observed (slight underestimation). In both studies, under and overestimation was slight (< 15% in the current study). We note that the filters used in the UPAS in both our study and the PURE study are no longer commercially available; however, better performance, especially at low concentrations, is anticipated with alternative and commonly available PTFE filters. This difference in filter performance is likely due to adsorption of semi-volatile aerosol that can occur on the fibrous filters (Kirchstetter et al., 2001) used in both this study and in the PURE study. At the high levels of mass deposition encountered in households that use biomass fuel, the overall impact of this adsorption may be relatively low; however, as households transition to cleaner fuels, it may substantially bias estimates upwards (Clark et al., 2013). Finally, the PURE study utilized an earlier version of the UPAS (version 1) compared to Version 2 used in our study. The second version of the UPAS enables use of PTFE filters with support rings to help minimize the aforementioned adsorption effect and enables user-defined duty cycles to extend battery life and, therefore, the duration over which samples are gathered (Arku

Figs. 2 and 3 suggest the potential for differential bias, with a larger underestimation of $PM_{2.5}$ exposures by the UPAS at higher concentrations. However, data at higher concentrations are sparse; it is thus not possible to determine if the potential differential bias is real nor to attribute the measurement error to the performance of the UPAS or to the performance of the pump and filter sampler.

Our study was limited by its sample size and its single geographic setting, where we observed a fairly narrow range of $PM_{2.5}$ exposures. Our evaluation occurred over a relatively short time period in our Western Honduras study population, and thus did not present an opportunity to evaluate the performance of the UPAS across multiple seasons or locations that would represent more variability in temperature and humidity. A strength of our study was the opportunity to observe and report on field performance of a new monitor for personal PM exposure assessment. For example, during the initial phase of our evaluation, we visually inspected the filters and observed poor correlations between the UPAS and other gravimetric samplers (data not shown). We determined this was likely due to large particles (i.e., larger than $PM_{2.5}$) accumulated over time in the grit pot of the UPAS cyclone and subsequently deposited erroneously on the filter. This issue, which leads to overestimation of mass deposition, may have been anticipated

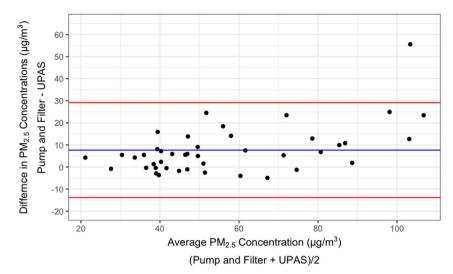


Fig. 3. Bland-Altman plot comparing differences in paired 24-hour personal $PM_{2.5}$ concentrations (y-axis) and the averages of the paired $PM_{2.5}$ concentrations (x-axis) collected with two different measurement techniques (UPAS and the Pump and Filter; n=43). The blue line represents the mean of the differences; the red lines represent the 95% confidence interval of the differences. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

given the orientation of the UPAS when worn. The issue was resolved by applying high-vacuum grease to the grit pot of the UPAS cyclone and with regular cleaning and replacement of grease (Access Sensor Technologies, 2018). While standard protocols for UPAS deployment now account for this issue, past experience has shown that new measurement devices often fail in unexpected ways. As development of new air pollution sensing technologies accelerates, we expect that transparent documentation of device performance, as presented here, would contribute to standard protocol development for laboratory and field performance testing and advance exposure science.

The UPAS has a number of other features – including GPS – and continues to be developed, including on-going work on a semi-continuous measure of $PM_{2.5}$ concentration based on post hoc analyses of monitored and recorded pressure changes across the filter as mass accumulates. While we did not use these during this evaluation, they are likely to be of value to other researchers, and use and evaluation of these features is encouraged for future studies. Further evaluation of the UPAS for personal exposure measurements across a wider range of geographic settings with more diverse air pollution sources, including other solid fuels, traffic-related air pollution, dust, and industrial sources, among others, will expand the evidence base for UPAS performance. Additional performance parameters – such as noise levels and participant feedback on the monitor – should also be assessed in future evaluations.

Unlike past assessments, our study focused on collocated personal samples, rather than stationary measures, and provided insights that refined the instrument's standard operating procedure. Twenty-four hour average concentrations in our study were up to 130 µg/m³. Although personal household air pollution exposure concentrations have been reported 2- to 5-fold higher than this in other geographic settings with solid fuel use (between 220 and $900 \,\mu g/m^3$ (Pope et al., 2017) in a recent review of interventions), our study still covered a range of PM_{2.5} concentrations that is relevant to many settings worldwide (Health Effects Institute, 2018). Our findings provide strong, albeit preliminary, evidence that the UPAS is a suitable monitor for measuring personal exposures to household air pollution. Given its weight, size, battery life, relative silence, and performance, the UPAS represents a viable option for exposure assessment in settings where households use solid fuels to meet their daily energy needs and, potentially, for numerous other environments in which PM_{2.5} exposure is a concern.

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