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Kitchen Concentrations of Fine Particulate Matter and Particle Number Concentration in Households Using Biomass Cookstoves in Rural Honduras

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Abstract

Cooking and heating with solid fuels results in high levels of household air pollutants, including particulate matter (PM); however, limited data exist for size fractions smaller than PM2.5 (diameter less than 2.5 µm). We collected 24-h time-resolved measurements of PM2.5 (n = 27) and particle number concentrations (PNC, average diameter 10–700 nm) (n = 44; 24 with paired PM2.5 and PNC) in homes with wood-burning traditional and *Justa* (i.e., with an engineered combustion chamber and chimney) cookstoves in rural Honduras.

The median 24-h PM2.5 concentration (n = 27) was 79 µg/m3 (interquartile range [IQR]: 44–174 µg/m3); traditional (n = 15): 130 µg/m3 (IQR: 48–250 µg/m3); *Justa* (n = 12): 66 µg/m3 (IQR: 44–97 µg/m3). The median 24-h PNC (n = 44) was 8.5 × 104 particles (pt)/cm3 (IQR: 3.8 × 104–1.8 × 105 pt/cm3); traditional (n = 27): 1.3 × 105 pt/cm3 (IQR: 3.3 × 104–2.0 × 105 pt/cm3); *Justa* (n = 17): 6.3 × 104 pt/cm3 (IQR: 4.0 × 104–1.2 × 105 pt/cm3). The 24-h average PM2.5 and particle number concentrations were correlated for the full sample of cookstoves (n = 24, Spearman ρ: 0.83); correlations between PM2.5 and PNC were higher in traditional stove kitchens (n = 12, ρ: 0.93) than in *Justa* stove kitchens (n = 12, ρ: 0.67). The 24-h average concentrations of PM2.5 and PNC were also correlated with the maximum average concentrations during shorter-term averaging windows of one-, five-, 15-, and 60-min, respectively (Spearman ρ: PM2.5 [0.65, 0.85, 0.82, 0.71], PNC [0.74, 0.86, 0.88, 0.86]).

Given the moderate correlations observed between 24-h PM2.5 and PNC and between 24-h and the shorter-term averaging windows within size fractions, investigators may need to consider cost-effectiveness and information gained by measuring both size fractions for the study objective. Further evaluations of other stove and fuel combinations are needed.

Keywords

Household air pollution, biomass, cookstoves, particle number concentration, solid fuel, Ultrafine particles, Real-time measurements

Disciplines

Environmental Public Health | Other Medicine and Health Sciences | Public Health

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Kitchen concentrations of fine particulate matter and particle number concentration in households using biomass cookstoves in rural Honduras

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Keywords: household air pollution; solid fuel; particulate matter; ultrafine particles; real-time measurements

Abstract

Cooking and heating with solid fuels results in high levels of household air pollutants, including particulate matter (PM); however, limited data exist for size fractions smaller than $PM_{2.5}$ (diameter less than 2.5 µm). We collected 24-hour time-resolved measurements of $PM_{2.5}$ (n=27) and particle number concentrations (PNC, average diameter 10-700 nm) (n=44; 24 with paired $PM_{2.5}$ and PNC) in homes with wood-burning traditional and *Justa* (i.e., with an engineered combustion chamber and chimney) cookstoves in rural Honduras.

The median 24-hour $PM_{2.5}$ concentration (n=27) was 79 µg/m^3 (interquartile range [IQR]: 44 – 174 µg/m^3); traditional (n=15): 130 µg/m^3 (IQR: 48 – 250 µg/m^3); *Justa* (n=12): 66 µg/m^3 (IQR: 44 – 97 µg/m^3). The median 24-hour PNC (n=44) was 8.5 x 10^4 particles (pt)/cm^3 (IQR: 3.8 x 10^4 – 1.8 x 10^5 pt/cm^3); traditional (n=27): 1.3 x 10^5 pt/cm^3 (IQR: 3.3 x 10^4 – 2.0 x 10^5 pt/cm^3); *Justa* (n=17): 6.3 x 10^4 pt/cm^3 (IQR: 4.0 x 10^4 – 1.2 x 10^5 pt/cm^3). The 24-hour average $PM_{2.5}$ and particle number concentrations were correlated for the full sample of cookstoves (n=24, Spearman ρ: 0.83); correlations between $PM_{2.5}$ and PNC were higher in traditional stove kitchens (n=12, ρ: 0.93) than in *Justa* stove kitchens (n=12, ρ: 0.67). The 24-hour average concentrations of $PM_{2.5}$ and PNC were also correlated with the maximum average concentrations during shorter-term averaging windows of one-, five-, 15-, and 60-minutes, respectively (Spearman ρ: $PM_{2.5}$ [0.65, 0.85, 0.82, 0.71], PNC [0.74, 0.86, 0.88, 0.86]).

Given the moderate correlations observed between 24-hour $PM_{2.5}$ and PNC and between 24-hour and the shorter-term averaging windows within size fractions, investigators may need to consider cost-effectiveness and information gained by measuring both size fractions for the study objective. Further evaluations of other stove and fuel combinations are needed.

Main Findings: Kitchen concentrations of fine particulate matter ($PM_{2.5}$) and particle number concentration were moderately correlated between traditional and improved biomass cookstoves.

1. Introduction

Approximately three billion people, predominantly in low- and middle-income countries, rely on solid fuels (e.g., wood, charcoal, dung) as their primary energy source for cooking (Bonjour et al., 2013). Combustion of solid fuels often results in levels of household air pollution that exceed World Health Organization (WHO) air quality guidelines (e.g., 24-hour mean $PM_{2.5}$ concentrations greater than 25 $\mu g/m^3$) (Thomas et al., 2015; World Health Organization, 2006a). This household air pollution is one of the top environmental risk factors for the global burden of disease and was estimated to be responsible for 1.6 million deaths and 59 million disability-adjusted life years in 2017 (Stanaway et. al, 2018).

Human exposure to particulate matter air pollution is typically assessed using gravimetric (i.e., mass-based) sampling of fine particulate matter ($PM_{2.5}$). The smallest particles, especially those smaller than 0.1 μm (i.e., ultrafine particles), may have important health implications. Ultrafine particles can penetrate deep into the lungs resulting in oxidative stress and systemic inflammation (Brauer et al., 2001; Brauner et al., 2007; Brook et al., 2010; Donaldson et al., 2001; Donaldson and Stone, 2003; Sioutas et al., 2005). Ultrafine particles have a high surface area, but very little mass, thus measuring these size fractions via gravimetric sampling is not suitable. As a result, particle number concentration (PNC) is a more relevant metric for ultrafine particles than mass concentration.

Several studies have explored the size distribution of ultrafine particles emitted from different cookstove technologies in the laboratory setting (Rapp et al., 2016; Shen et al., 2017; Tiwari et al., 2014; Tryner et al., 2018). Measuring concentrations of $PM_{2.5}$ and PNC in kitchens using traditional or engineered cookstoves in the field setting is logistically challenging and inhibited by monetary barriers associated with monitoring ultrafine particles. To date, only a few studies have used portable monitors to compare concentrations of $PM_{2.5}$ and particle number in kitchens using three-stone fires and other biomass cookstoves (Chowdhury et al., 2012; de la Sota et al., 2018; Eilenberg et al., 2018; Wangchuk et al., 2015; Zhang et al., 2012). There is currently no standard protocol for measuring PNC in household air pollution research, and results from field studies vary substantially due to variation in the instrumentation, sample duration, stove type evaluated, and sample sizes.

Generally, it is assumed that engineered cookstoves (i.e., those designed with the intent to burn fuel more efficiently) reduce indoor particulate matter mass, but the resulting changes in particle size are less clear. Risk assessments for household air pollution, to date, are often focused on $PM_{2.5}$ mass exposure and do not account for ultrafine PNC (Armendáriz-Arnez et al., 2010; Jetter et al., 2012; Smith et al., 2014). Laboratory studies suggest that certain engineered cookstoves (such as a natural draft top-lit up-draft stove) emit fewer particles in the ultrafine range (<0.1 μm) compared to traditional cookstoves (such as a three-stone fire) (Jetter et al., 2012); other engineered cookstoves (e.g., rocket and forced-draft gasifier cookstoves) demonstrate a shift to higher numbers of smaller particles (<0.03 μm) despite substantially reducing emissions of $PM_{2.5}$ mass (Jetter et al., 2012; Just et al., 2013; Rapp et al., 2016). Given the inconsistencies observed in the relationship between emissions of $PM_{2.5}$ mass and ultrafine particles during laboratory testing of various engineered biomass cookstove technologies, the estimated health benefits of engineered cookstoves may be misstated if field studies do not account for ultrafine particles.

The *Justa* cookstove is a commonly used engineered cookstove in Latin America and features an insulated, rocket-elbow combustion chamber, chimney, side compartment to remove excess ash, and *plancha* (Figure 1). Laboratory tests of the *Justa* stove show PM reductions of approximately one third compared to traditional three stone fires (Still et al., 2012). Additionally, a field study in Honduras demonstrated that the emission factor for improved stoves with chimneys was approximately 50% lower than that for traditional cookstoves (4.5 g kg vs. 8.2 g kg) (Roden et al., 2009). A study among 59 households in Honduras also showed that kitchens with *Justa* cookstoves had *73*% lower $PM_{2.5}$ concentrations compared to kitchens with traditional stoves (Clark et al., 2010). A field test of 5 kitchens in Honduras found that the *Justa* cookstove had lower $PM_{2.5}$ emissions factors than the traditional stove and that the geometric mean particle diameter was 48 nm (Eilenberg et al., 2018). To date, no field studies have examined PNC in kitchens using *Justa* cookstoves.

Although time-integrated gravimetric sampling is most often used to assess household air pollution exposure, time-resolved measurements provide additional insight into $PM_{2.5}$ concentrations with respect to temporal variability or intensity of exposure during and between cooking events (Carter et al., 2016; Chen et al., 2016; Ezzati et al., 2000a; Fischer and Koshland, 2007; Northcross et al., 2015; Park and Lee, 2003; Van Vliet et al., 2013). For example, Van Vliet et al. (2013) reported that a few short-term periods of elevated $PM_{2.5}$ concentrations constituted a substantial portion of daily exposure. It is unclear, however, whether metrics evaluated over periods shorter than 24-hours ("shorter-term" metrics) such as one-hour average maximum concentrations, may be relevant for health models evaluating effects of household air pollution.

In this study, we used real-time instrumentation to quantify kitchen concentrations of $PM_{2.5}$ and PNC in rural areas surrounding La Esperanza, Honduras where biomass (wood-fueled) cookstoves were primarily used for cooking. Our objective was to evaluate and compare wood-burning traditional and *Justa* cookstoves (the latter of which had an engineered combustion chamber and chimney). Our goals were to 1.) characterize real-time $PM_{2.5}$ mass concentrations and real-time PNC, 2.) evaluate the correlations between 24-hour average $PM_{2.5}$ mass and particle number concentrations (for all stoves and by stove type), and 3.) evaluate correlations between 24-hour average concentrations and shorter-term averaging windows for both pollutants. To our knowledge, our study is the first household air pollution study to measure paired 24-hour real-time concentrations of $PM_{2.5}$ and PNC.

2. Materials and Methods

2.1 Study Site, Population, and Stove Types

This study was conducted in rural communities surrounding La Esperanza, Department of Intibucá, Honduras as part of a larger study evaluating the health effects of exposure to household air pollution. In brief, the larger study included 230 women, aged 24-59, who were non-smokers and not pregnant. We measured real-time $PM_{2.5}$ and PNC in a subsample of the women's kitchens. With only one set of monitoring equipment, we were limited to collecting data from one kitchen per day. We collected forty-seven 24-hour samples in 36 unique kitchens from August 2015 to December 2016. We used a household survey to assess physical characteristics of the kitchen. We recorded the number of walls, windows, and doors; kitchen volume (height x length x width); wall material (mud, sticks); floor material (concrete, dirt, tile); roof material (sheet metal or tile); and presence of eaves. Additionally, women self-reported their use of a secondary stove as well as the number of cooking events and the number of people they cooked for during the 24-hour monitoring period (Young et al., 2019).

Of the 47 samples collected, 30 were collected in households that used a traditional cookstove and 17 were collected in households that used a *Justa* cookstove. Traditional cookstoves were typically self-built adobe stoves, with a metal *plancha* (griddle), a non-insulated open combustion area, and sometimes a chimney (Kshirsagar and Kalamkar, 2014; Kumar et al., 2013). All *Justa* stoves were installed in the homes approximately six months prior to the measurements. Stove users reported burning gathered wood, including split logs and sticks, as the primary fuel in both cookstoves. Additionally, users reported burning small sticks of a local wood called ocote (a species of pine) and corncobs to start the fire.

2.2 Particle Measurements

2.2.1 Fine particulate matter ($PM_{2.5}$)

$PM_{2.5}$ was sampled using an aerosol nephelometer, the personal DataRam (pDR) 1200 (Thermo Fisher Scientific Inc., Waltham MA, USA), powered by a 9V lithium ion rechargeable battery. The pDR was set up in an active-flow mode (1.5 L/min) using a pump (SKC AirChek XR5000 pump) and $PM_{2.5}$ cyclone inlet (Triplex Cyclone; Mesa Labs, Butler NJ, USA). A 37mm filter (Fiberfilm™, Pall Corporation, Port Washington NY, USA) was installed downstream of the pDR photometric sensing chamber. The setup enabled estimation of time-resolved (60-second averaged) $PM_{2.5}$ mass concentration followed by (downstream) collection of a time-integrated gravimetric sample. We collected field blanks once a week. An external data logger (EasyLog EL-

USB-2, Lascar Electronics Ltd., Erie PA, USA) recorded the one-second pDR analog voltage data (0-5 V), corresponding to $PM_{2.5}$ concentrations between 0 and 4,000 μg/m^3. The pDR was zeroed in ambient air and the triplex cyclone was thoroughly cleaned before each 24-hour sample. At the field house in La Esperanza, the pump flow rate was checked pre- and post-sample using a flow meter (Bios International DryCal Lite, Mesa Labs, Butler NJ, USA). Sample filters were stored in a -20°C freezer in Honduras until they were transported back to Colorado State University and stored in a -80°C freezer. All filters were pre- and post-weighed to the nearest microgram (Mettler Toledo MX5, Mettler OH, USA) at Colorado State University, USA. Filters were equilibrated for 24 hours prior to weighing. Filter mass was determined by weighing each filter twice and averaging the weights. If the weights differed by more than 5 μg, a third weight was taken and the average of all three was used.

2.2.2 Ultrafine PNC

PNC was measured with the DiSCMini (Testo AG, Germany; Fierz et al., 2011). The DiSCMini is a handheld diffusion size classifier that estimates particle number for particles with diameters between 10 and 700 nm and provides data on airborne PNC between 10^3 and 10^6 particles (pt)/cm^3. Portable, direct reading instruments, such as the DiSCmini, are a relatively new technology for field and personal monitoring of particle number. In both laboratory and field tests, the DiSCMini demonstrates high correlation with other instruments that measure particle number concentration (Aerotrack 9000, P-TRAK, and scanning mobility particle sizer [SMPS]) when tested in the same settings, indicating that the DiSCMini is a useful instrument for field monitoring (Asbach et al., 2012; Bau et al., 2017; Fierz et al., 2009a; Meier et al., 2013; Mills et al., 2013; Viana et al., 2015). The instrument has been shown to report within ± 30% for mean particle size and number concentration (Asbach et al., 2012). We equipped the DiSCMini with an external rechargeable battery to ensure 24 hours of continuous monitoring (7.4V 7.8Ah custom lithium ion battery). The DiSCMini recorded and logged concentrations at 1 Hz. The impactor on the DiSCMini inlet (cutpoint = 0.7 μm) was thoroughly cleaned before each 24-hour sample to help maintain flow through the instrument. The DiSCMini instrument turns off the pump for one minute in every hour to measure the zero offset in order to account for any long-term drifts in temperature or humidity (Fierz et al., 2011) .

2.2.3 Household-level Field Measurements

The DiSCMini and pDR were collocated 40 to 70 inches from the front edge of the stove, 42 to 95 inches above the ground, and 41 to 61 inches from the nearest wall in each kitchen (Figure 2). Both instruments were started manually. The pump for the active $PM_{2.5}$ measurements was programmed to turn off after 24 hours; the DiSCmini was manually switched off after 24 hours. A temperature and relative humidity monitor with a 60-second resolution (EasyLog EL-USB-2, Lascar Electronics Ltd., Erie PA, USA) was also collocated with the pDR and DiSCMini (Figure 2).

2.3 Data Processing

2.3.1 Fine particulate matter ($PM_{2.5}$)

The real-time pDR measurements below the limit of detection (LOD) of 5 µg (Wallace et al., 2011) were substituted with the LOD/(√2) (Hewett and Ganser, 2007). Real-time pDR measurements were then corrected for relative humidity using Equation 1 (Chakrabarti et al., 2004):

$$PM_{2.5,60-s,dry} = \frac{PM_{2.5,60-s,wet}}{1 + 0.25RH^2/(1 - RH)} \quad (1)$$

where $PM_{2.5,60-s,dry}$ was the dry (i.e., RH-corrected) 60-second average $PM_{2.5}$ concentration, $PM_{2.5,60-s,wet}$ was the 60-second average $PM_{2.5}$ concentration recorded by the pDR, and RH was the relative humidity. In addition, we normalized real-time pDR concentrations to gravimetric measurements as shown in Equation 2:

$$PM_{2.5,60-s,corr} = \frac{PM_{2.5,60-s,dry}}{\left(PM_{2.5,pDR}/PM_{2.5,filter}\right)_{24-hour}} \quad (2)$$

where $PM_{2.5,60-s,corr}$ was the LOD-, RH-, and filter-corrected 60-s average $PM_{2.5}$ concentration, $PM_{2.5,60-s,dry}$ was the LOD- and RH-corrected 60-s average $PM_{2.5}$ concentration, and the denominator is the LOD- and RH- corrected 24-hour average $PM_{2.5}$ concentration measured using the pDR ($PM_{2.5,pDR}$) divided by the 24-hour average $PM_{2.5}$ concentration measured using the filter ($PM_{2.5,filter}$). The value of$PM_{2.5,filter}$, in μg/m^3, was calculated from the mass accumulated on the filter (corrected for 25 filter blanks and the LOD), the sample duration, and average of the pre- and post-test flow rates. The $PM_{2.5}$ mass LOD was calculated by adding the average mass of the field blanks to three times the standard deviation of field blank masses (MacDougall et al., 1980). Filter weights below the LOD were substituted with the LOD/√2 (Hewett and Ganser, 2007).

2.3.2 Particle Number Concentration

The data from the DiSCmini were preprocessed using the DiSCmini data conversion tool (Matter Aerosol 2011, version 2.0), which assumes the number median diameter on the diffusion and filter stages was 30 nm and 300 nm, respectively. All additional analyses were performed in R, version 3.4.1 (R Core Team, Vienna, Austria). Given that the DiSCMini monitor was prone to overloading due to high emissions from the cookstoves and poor ventilation in the kitchen, we checked the DiSCmini data log for each household measurement for various error codes for each second sampled. For example, the DiSCmini electrometer amplifiers can detect currents between zero and 4096fA. At very high particle concentrations, the electrometer amplifiers will reach their maximal level and produce an error code. Additional errors can occur due to large temperature variations, high relative humidity, dirt on the charger's corona wire, or flow of the instrument falling below 0.95 liters per minute. Rapid changes in particle concentrations can also result in negative diffusion and filter stages (Fierz et al., 2009b). Of the 3,801,600 total seconds in our dataset, <1% of the data had at least of one of the following error codes: filter stage below zero, diffusion stage below zero, filter stage or diffusion stage over 4096 fA (total current). All seconds flagged with an error code were excluded from the data analyses. Following the removal of seconds flagged with errors, we aggregated data to one-minute intervals using the mean PNC of each sampling minute.

2.4 Data Analysis

2.4.1. Goal 1: Characterize $PM_{2.5}$ and PNC

We calculated descriptive statistics for the samples from the one-minute averages for both the $PM_{2.5}$ (LOD-, RH- and filter-corrected) and PNC data sets. We calculated the 24-hour minimum, maximum, mean, median, standard deviation, 25th and 75th percentiles, as well as maximum concentrations in one-minute, five-minute, 15-minute, and 60-minute moving windows within each kitchen. We also created descriptive plots of the 24-hour real-time concentrations of $PM_{2.5}$ and PNC for each kitchen. We used the pDR data ($PM_{2.5,60-s,corr}$) to compute the number of minutes that each sample's $PM_{2.5}$ concentration was above 100 μg/m^3 (the equivalent of four times the WHO 24-hour air quality guideline) (World Health Organization, 2006a); a metric previously observed to be associated with increased incidence of acute lower respiratory infections among children (Chen et al., 2016; Gurley et al., 2013). A nonparametric alternative to the t-test, Wilcoxon rank sum test, was used to test for differences in 24-hour average $PM_{2.5}$, 24-hour average PNC, and number of hours spent above 100 μg/m^3 by stove type. Finally, for each sample, we removed 60-s average $PM_{2.5}$ concentrations above the sample's 95th percentile and then re-calculated the 24-hour average sample $PM_{2.5}$ concentration (without the top 5th percentile) to evaluate the contribution of these high-concentration periods on the 24-hour kitchen concentration.

2.4.2 Goal 2: Correlation between PNC and $PM_{2.5}$

We calculated Spearman correlation coefficients (a non-parametric test used due to non-normally distributed data) between the following data: 1.) 24-hour $PM_{2.5}$ and PNC, 2.) maximum one-hour average $PM_{2.5}$ and PNC, and 3.) maximum one-minute $PM_{2.5}$ and PNC for all households and by stove type. Since the number of air changes within the houses may affected the correlation between 24-hour $PM_{2.5}$ and PNC, we conducted a sensitivity analysis to characterize correlations between $PM_{2.5}$ and PNC for samples above and for those below the median number of air exchanges per hour for the sample. We used real-time $PM_{2.5}$ data from the pDR to calculate the number of air changes per hour for 23 households. For each household, we selected a single decay event where the $PM_{2.5}$ concentration reached a peak and then fell continuously to a lower concentration. We made sure to select a decay event that consisted of at least 15 minutes of data. We then fit the data to a linear model using ordinary least squares regression. The time since the maximum concentration occurred, t, was the independent variable and $\ln(c/c_i)$ was the dependent variable (where c was the $PM_{2.5}$ maximum concentration and c_i was the concentration at time in hours since the maximum concentration occurred). The absolute value of the model slope was described as the number of air exchanges per hour (AEPH) (Burgess et al., 2004).

2.4.3 Goal 3: Shorter-Term Concentrations

We calculated Spearman correlation coefficients between all averaging windows (24-hour, maximum one-minute, maximum five-minute, maximum 15-minute, and maximum 60-minute) within both $PM_{2.5}$ and PNC.

3. Results

Of the forty-seven 24-hour samples performed, we excluded 20 $PM_{2.5}$ samples (16 samples with external data logger failures, three with missing temperature and humidity data, one with negative gravimetric data). We excluded three PNC samples (all from homes using traditional stoves) because the DiSCMini turned off prior to completing at least 80% of the 24-hour sampling period. Our final sample size was 27 $PM_{2.5}$ samples (collected in 24 unique kitchens) and 44 PNC samples (collected in 36 unique kitchens).

Kitchen characteristics of the sample population are described in Table 1. The majority of kitchens were constructed of mud or stuccoed adobe walls (60%), with dirt floors (70%) and sheet metal roofs (70%). Approximately 30% of all households reported having a traditional secondary stove occasionally used for cooking. In Honduras, secondary stoves are typically used outside the home for cooking large pots of beans or corn. During the 24-hour monitoring period, women reported cooking a mean of 3.1 times (SD: 0.88 times) for a mean of 5.5 people (SD: 2.5 people). The median number of air exchanges per hour was 9.7 (mean: 10.5; range: 3.6 to 18.2).

3.1 Goal 1: Characterize $PM_{2.5}$ and PNC

The median gravimetrically-determined 24-hour average $PM_{2.5}$ concentration for all samples (n=27) was 79 μg/m^3 (IQR: 44-174 μg/m^3) (Table 2). On average, households using traditional primary stoves had higher $PM_{2.5}$ concentrations (median: 130 μg/m^3; IQR: 48-250 μg/m^3; n=15) compared to households using *Justa* stoves (median: 66 μg/m^3; IQR: 44-97 μg/m^3; n=12) (Wilcoxon rank sum test, p = 0.11) (Table 2). The average ratio of the $PM_{2.5}$ concentrations measured using the pDR and the filter [$(PM_{2.5,pDR}/PM_{2.5,filter})_{24-hour}$; the "response factor"] was 0.56 (median: 0.57), indicating that the nephelometer tended to underestimate $PM_{2.5}$ concentrations relative to the time-integrated filter measurements (IQR: 0.39-0.66; n=27). The median pDR response factors by stove type were 0.60 (IQR: 0.45-0.74; n=15) for traditional stoves and 0.50 (IQR: 0.37-0.58; n=12) for *Justa* cookstoves.

During the one-hour averaging windows, maximum $PM_{2.5}$ concentrations ranged from 51 to 4026 μg/m^3 for all 27 samples, 141 to 4026 μg/m^3 for traditional stoves, and 51 to 2098 μg/m^3 for *Justa* stoves (Figure 3). On average, one-hour maximum concentrations were higher for traditional stoves (mean: 1469 μg/m^3; SD: 1141 μg/m^3; n=15), compared to *Justa* stoves (mean: 957 μg/m^3; SD: 719 μg/m^3; n=12; Wilxcon rank sum, p= 0.32; Figure 3). The average number of hours a kitchen $PM_{2.5}$ concentration exceeded 100 μg/m^3 was 4.0 hours (SD: 3.7; n=27) and ranged from less than 1 hour to over 15 hours. Kitchen $PM_{2.5}$ concentrations exceeded 100 μg/m^3 for a mean of 5.5 hours (SD: 4.4; n=15) in kitchens with traditional cookstoves and a mean of 2.3 hours (SD: 1.3; n=12) in kitchens with *Justa* stoves (Wilcoxon p=0.08) (Table 3).

When corrected 60-s average $PM_{2.5}$ concentrations ($\boldsymbol{PM_{2.5,60-s,corr}}$) above the 95th percentile were removed, the median 24-hour $PM_{2.5}$ concentration in kitchens was 25 μg/m^3 (IQR: 15-62 μg/m^3; n=27) (Traditional stoves: 50 μg/m^3; IQR: 16-127 μg/m^3; n=15; *Justa* stoves: 19 μg/m^3; IQR: 15-30 μg/m^3; n=12; Wilcoxon rank sum, p=0.14). On average, the 60-second concentrations above the 95th percentile exposure values accounted for 42% of the 24-hour average concentration (46% among traditional stoves and 37% among *Justa* stove). The Spearman rho correlation between the full 24-hour average $PM_{2.5}$ concentration dataset and the dataset with the top 5% removed was 0.90. The Spearman rho correlation between the full 24-hour average $PM_{2.5}$ concentration and the dataset with only concentrations above the 95th percentile was 0.96.

The 24-hour average mean and median PNC for 44 samples are shown in Figure 4; the median concentration was 8.5x10^5 pt/cm^3 (IQR: 3.8x10^4- 1.8x10^5 pt/cm^3; n=44). PNC was lower among the households with *Justa* cookstoves (median: 6.3x10^4 pt/cm^3; IQR: 4.0x10^4-1.2x10^5 pt/cm^3; n=17) compared to traditional cookstoves (median: 1.3x10^5 pt/cm^3; IQR: 3.3x10^4-2.0x10^5 pt/cm^3; n=27); however, the pollutant distributions largely overlapped for the two stove types (Wilcoxon rank sum, p = 0.76). Descriptive plots of the 24-hour concentrations for $PM_{2.5}$ mass and PNC for individual kitchens demonstrate similar patterns in PNC and $PM_{2.5}$ emissions throughout the day (example presented in Figure 5).

3.2 Goal 2: Correlation between $PM_{2.5}$ and PNC

The Spearman correlation coefficient between filter-corrected 24-hour average $PM_{2.5}$ and 24-hour average PNC was 0.83 (n=24). Correlations between 24-hour average $PM_{2.5}$ and PNC were higher in traditional stove households (ρ=0.93) than in *Justa* stove households (ρ=0.67). The Spearman correlation between the maximum one-hour-average $PM_{2.5}$ and the maximum one-hour-average ultrafine PNC was 0.54 (traditional stoves: ρ=0.62; *Justa* stoves: ρ=0.43, while the correlation between maximum one-minute concentrations of these two pollutants was 0.43 (traditional stoves: ρ=0.83; *Justa* stoves: ρ=0.36). The Spearman correlations between the number of air exchanges per hour and the 24-hour average pollutant concentrations were low ($PM_{2.5}$: ρ = 0.02; PNC: ρ = -0.08). Similarly low correlations between number of AEPH and 24-hour average concentrations were observed when each stove type was considered individually (traditional: $PM_{2.5}$: ρ = 0.17; PNC: ρ = 0.01; *Justa*: $PM_{2.5}$ ρ = -0.12; PNC ρ = -0.16). Spearman correlations between $PM_{2.5}$ and PNC above and below the median AEPH were high and similar between the two metrics (above the median: ρ = 0.82; below the median: ρ = 0.79).

3.3 Goal 3: Shorter-Term Concentrations

The maximum $PM_{2.5}$ concentrations for the one-minute, five-minute, 15-minute and 60-minute averaging periods were highly correlated with the 24-hour average for the household (ρ ranging from 0.65-0.85). Correlations between maximum values in one-minute, five-minute, and 60-minutes ranged from 0.58-0.95. Maximum PNC concentrations for the one-minute, five-minute, 15-minute, and 60-minute averaging windows were also highly correlated with the 24-hour average for the household (ρ ranging from 0.74-0.88).

4. Discussion

4.1 Goal 1: Characterize $PM_{2.5}$ and PNC

4.1.1 $PM_{2.5}$

Kitchen concentrations of $PM_{2.5}$ were higher among households with traditional stoves compared to kitchen concentrations where *Justa* stoves were used; however, there were substantial overlaps in average concentrations. Additionally, the mean 24-hour average $PM_{2.5}$ concentrations exceeded the World Health Organization (WHO) air quality guideline of 25 μg/m^3 for both cookstove types (World Health Organization, 2006b). Although our measures were for kitchen (area) and not personal concentrations, only 3 of 27 (22%) kitchens had 24-hour average concentrations below 25 μg/m^3 (one traditional, two *Justa*).

We observed variation over each 24-hour sampling period. Indoor $PM_{2.5}$ mass and ultrafine particle concentrations peaked in the morning, likely due to cookstove startup (generally between 4am-5am), and were lowest overnight when the stove was likely off. Similar studies using temporally-resolved emissions monitoring in Kenya and China have also observed elevated concentrations of $PM_{2.5}$ coinciding with diurnal patterns and phases of cooking (i.e., startup) (Carter et al., 2016; Ezzati et al., 2000a; Kaur et al., 2017; Park and Lee, 2003). The substantial variation in kitchen concentrations of $PM_{2.5}$ (within households) suggests that peaks of exposure occur during cooking (especially in scenarios without other primary sources of pollution, as with our study population) and highlight the importance of using personal monitoring to capture a better estimate of exposure (Clark et al. 2013).

4.1.2 PNC

In general, our results show lower PNC among the kitchens with a *Justa* cookstove, compared to traditional cookstove kitchens, despite substantial overlap in the concentrations. This result is similar to results reported in previous studies conducted in Senegal and China. De la Sota et al. (2018) used the DiSCmini to monitor PNC during cooking periods in three households using a traditional stove and three households using an improved rocket stove in Senegal. de la Sota et al. (2018) observed lower PNC during cooking events in the households with improved rocket stoves (median PNC 1.5×10^6 pt/cm^3) compared to the households with traditional stoves (median PNC 2.2×10^6 pt/cm^3). Similarly, a field study of 15 households using coal or wood for heating and cooking in China measured PNC with an AEROTRAK 9000 and reported that cookstoves with chimneys reduced kitchen concentrations of ultrafine particle by a factor of four during cooking periods (Hosgood et al., 2012). We did not calculate cooking event concentrations, which limits our ability to compare our results to both of these prior studies.

4.2 Goal 2: Correlation between PNC and $PM_{2.5}$

Correlation between 24-hour average $PM_{2.5}$ and PNC concentrations was high among traditional stoves (ρ=0.93) and moderate among improved stoves (ρ=0.67). The mechanism influencing the lower correlation of $PM_{2.5}$ mass and PNC in households with the *Justa* stove, compared to households with traditional stoves, is unclear and may be driven by differences in particle formation and oxidation processes resulting from differences between the two stove designs. The lower correlation could also be due to measurement error or influenced by a small number of *Justa* households sampled. Several *Justa* cookstove kitchens had high $PM_{2.5}$ concentrations with relatively low PNC; these samples, in particular, may have been subject to measurement error. The 0.7 µm impactor on the DiSCmini inlet was occasionally clogged by large particles in the high-concentration kitchen setting, resulting in reduced flow through the instrument.

The correlations between $PM_{2.5}$ and PNC above and below the median number of air exchanges per hour were similar. We observed low correlations between the 24-hour average concentrations and the number of air exchanges per hour. One might expect 24-hour average pollutant concentrations to be negatively correlated with the number of air changes per hour, since higher air exchange rates would help remove cookstove emissions from the home; however, we observed both low positive and low negative correlations. One may also expect the correlations between the number of air exchanges per hour and concentrations to be higher, however we did not ask participants to keep a log of activities and it could be possible that changes in behavior such as opening or closing windows and doors.

For both stove types combined, correlation between maximum one-hour average $PM_{2.5}$ mass and PNC was moderate (ρ=0.64) and correlation between one-minute maximum $PM_{2.5}$ mass and PNC was lower (ρ=0.41). These results indicate possible differences in exposure to $PM_{2.5}$ and ultrafine PNC over short time periods, perhaps during transient operating conditions that occur when the fire is started or refueled. Evidence from both lab and field studies support the hypothesis that variations in particle size and PNC are related to certain cooking activities and phases of cooking (Arora et al., 2013; Carter et al., 2016; Ezzati et al., 2000b; Park and Lee, 2003; Tryner et al., 2018; Zhang et al., 2012, 2010).

4.3 Goal 3: Shorter-Term Concentrations

Our one-hour average maximum $PM_{2.5}$ concentrations for individual samples ranged from 44 to 3929 µg/m^3 and were slightly lower than one-hour average concentrations reported by Fischer (159 to 6200 µg/m^3; n=43) among households using a variety of stoves in China (Fischer and Koshland, 2007). The highest $PM_{2.5}$ concentrations in one-hour averaging windows among traditional and *Justa* stove samples (3929 and 1682 µg/m^3, respectively) were 15 and 22 times higher than the 24-hour averages. Park and Lee observed peak kitchen concentrations (defined as at least 7 minutes before pollutant decay was observed) between 32 and 39 times higher than 24-hour averages among traditional and improved biomass stove users in Costa Rica (Park and Lee, 2003). The mean sample one-hour average maximum values were 207 and 87 times larger than baseline levels measured at night for traditional and *Justa* stove groups, respectively. Other studies suggest that high short-term particulate matter concentrations likely result from cooking activities such as tending or adding fuel to the fire (Bartington et al., 2017; Ezzati et al., 2000a; Just et al., 2013). Additionally, Van Vliet et al. observed that mitigating the overall highest 1-5% of the 60-second $PM_{2.5}$ concentrations during a 24-hour sampling period in a Ghana field study could reduce mean personal 24-hour exposure by 49-75% (Van Vliet et al., 2013). We observed that removing 60-second $PM_{2.5}$ real-time concentrations above the 95th percentile decreased the overall 24-hour average concentrations by 42%, 46% among traditional stoves and 37% among *Justa* stoves. Given the high contribution of the most polluted concentrations to the overall average, it may be important to understand how reductions in start-up and cooking-specific emissions from different stove designs could reduce overall average concentrations.

The mean number of hours spent over 100 µg/m^3 (4.0 hours; IQR: 1.5-5.2) in our study kitchens was similar to the mean number of hours spent over 100 µg/m^3 (mean: 5.3, IQR: 4.0-6.9) among both homes using clean fuels and homes using biomass fuels in Bangladesh (Gurley et al., 2013). The health implications of short-term high concentrations of PM mass or ultrafine PNC are unclear in the field of household air pollution. Ambient air pollution studies have observed the association of one-hour average maximum ambient $PM_{2.5}$ on hospital admission and mortality but this association has not been studied for household air pollution (Burgan et al., 2010). Although our measured short-term kitchen concentrations of $PM_{2.5}$ are highly correlated with 24-hour concentrations, we do not know if this relationship is similar for personal exposure. It may be useful to measure short-term intensity of personal exposure when studying exposure-response relationships, especially for cardiovascular endpoints.

4.4 Limitations and Lessons Learned

Our study is limited by a small sample size. Use of the DiSCmini in settings with very high particle concentrations over a long period was also challenging. The inability of the DiSCMini and analog function on the pDR to report concentrations above 1,000,000 particles/cm^3 and 4,000 μg/m^3, respectively, could have led to measurement errors that would affect the correlation between the one-minute maximum $PM_{2.5}$ and PNC values. True concentrations may be higher than reported; however only <1% of all PNC data was above the upper limit of detection. The pDR instrument is suitable for fieldwork and areas of high concentrations; however, we experienced frequent loss of pDR data. We suspect that data loss resulted from the connection between the pDR and external logger coming loose during transportation, setup, or operation. Although humidity levels in our sample were high, 82% of minutes had humidity levels of 62%, we believe humidity corrections for $PM_{2.5}$ and the internal corrections for the DiSCMini were adequate in addressing humidity concerns for the measurements. The correlation between minute-level humidity and corrected concentrations were low ($PM_{2.5}$ = 0.05; PNC = -0.18 for PNC).

For each household, we estimated the number of air changes per hour using pDR data recorded during a single decay event. More robust estimates might have been obtained by using data from multiple decay events. Our ability to identify multiple decay events for each household was limited because (a) we did not ask occupants to record the times of cooking events and (b) multiple short-term concentration peaks were observed during many of the presumed cooking events.

Placement of the collocated instruments in the kitchens varied (due to logistical challenges of placing the instrument away from the stove and windows or door) and this variation could have affected the measurements. There are currently no standards for measuring ultrafine particles in the household setting, and it is unclear as to how the distance between the stove and the instruments may affect individual kitchen concentrations (He et al., 2005). Given the lack of standards for consistent spacing between monitors and stoves for measuring ultrafine PNC, it is possible our 24-hour concentrations and correlations between $PM_{2.5}$ mass and PNC would not be generalizable to other populations with different cooking environments (i.e., kitchen layout, ventilation), fuels, and stove types. Finally, our results for kitchen concentrations of $PM_{2.5}$ and PNC may not translate to measurements of personal $PM_{2.5}$ and PNC.

5. Conclusions

This study is the first to characterize 24-hour time-resolved $PM_{2.5}$ and ultrafine particle number concentrations in kitchens. Our study reveals variability in $PM_{2.5}$ and PNC within and between samples and indicates that the highest exposure periods account for almost one half of the 24-hour average concentration. Correlations between $PM_{2.5}$ and PNC differed between traditional and *Justa* stoves, indicating that additional research may be needed to understand how the correlation between $PM_{2.5}$ mass and ultrafine particle concentrations differs by stove type. This information would provide insight regarding whether measurements of fine particulate matter are sufficient for characterizing exposure to household air pollution, particularly for studies evaluating multiple stove types. High correlations between 24-hour averages and sub-daily concentrations of $PM_{2.5}$ and PNC indicate that monitoring 24-hour average concentrations in similar rural settings may be a cost-effective method (i.e., without incorporating real-time instrumentation) to evaluating household-level concentrations of $PM_{2.5}$ and ultrafine particulate matter.

Figure 1: Left: Example of a traditional cookstove in the Honduran study homes. Right: Example of a Justa cookstove

Figure 2: Left: DiSCmini instrument used to measure ultrafine particle number concentration. Right: Example set-up of DiSCMini and pDR monitors.

Table 1: Kitchen characteristics of study homes in rural Honduras

	n*	Mean	SD
Kitchen volume (m^3)	36	35.4	15.4
Number of walls	36	4.0	0.0
Number of windows	36	1.1	0.7
Number of doors	36	1.6	0.7
Number of people cooked for in past 24 hours	47	5.5	2.5
Number of times cooked in past 24 hours	47	3.1	0.88
Number of Air Changes Per Hour**	23	10.47	3.92
		n	%
Stove type	47		
Traditional		30	64%
Justa (Improved)		17	36%
Wall material	36		
Mud (adobe)		13	36%
Stuccoed adobe		16	45%
Wood/sticks		4	11%
Concrete		3	8%
Floor material	36		
Dirt		24	65%
Concrete		10	27%
Ceramic tile		3	8%
Roof material	36		
Sheet metal		25	70%
Tiles		12	30%
Use of secondary stove	47		
Yes		12	30%

*A total of 47 samples were collected in 36 unique households; 11 houses had repeated measurements and household characteristics remained the same

**One of the real-time pDR samples could not be calculated for an air exchange per hour due to short decay rates less than 15 minutes

Table 2: 24-hour average fine particulate matter ($PM_{2.5}$) and particle number concentration (PNC) among kitchens in rural Honduras

		Mean	Standard Deviation	Min	25th Percentile	Median	75th Percentile	Max
$PM_{2.5}$ μg/m^{3}*	All samples (n=27)	180	301	11	44	79	174	1467
	Justa (n=12)	76	51	11	44	66	97	173
	Traditional (n= 15)	263	386	23	48	130	250	1468
PNC (pt/cm3)	All samples (n=44)	1.20E+05	1.00E+05	4.40E+02	3.80E+04	8.50E+04	1.80E+05	4.10E+05
	Justa (n=17)	9.10E+04	6.90E+04	2.50E+04	4.00E+04	6.30E+04	1.20E+05	2.40E+05
	Traditional (n=27)	1.30E+05	1.10E+05	4.40E+02	3.30E+04	1.30E+05	2.00E+05	4.10E+05

Reduced dataset for 24-hour samples that had both $PM_{2.5}$ and PNC

		Mean	Standard Deviation	Min	25th Percentile	Median	75th Percentile	Max
$PM_{2.5}$ μg/m^{3}*	All samples (n=24)	124	164	11	41	70	142	805
	Justa (n=12)	76	51	11	44	66	97	173
	Traditional (n= 12)	172	220	23	44	99	201	804
PNC (pt/cm3)	All Stoves (n=24)	9.50E+04	7.90E+04	1.90E+04	3.80E+04	6.20E+04	1.40E+05	2.60E+05
	Justa (n=12)	8.10E+04	7.10E+04	2.50E+04	3.80E+04	5.50E+04	8.50E+04	2.40E+05
	Traditional (n=12)	1.10E+05	8.60E+04	1.90E+04	3.80E+04	8.00E+04	1.60E+05	2.60E+05

***Corrected to gravimetric samples**

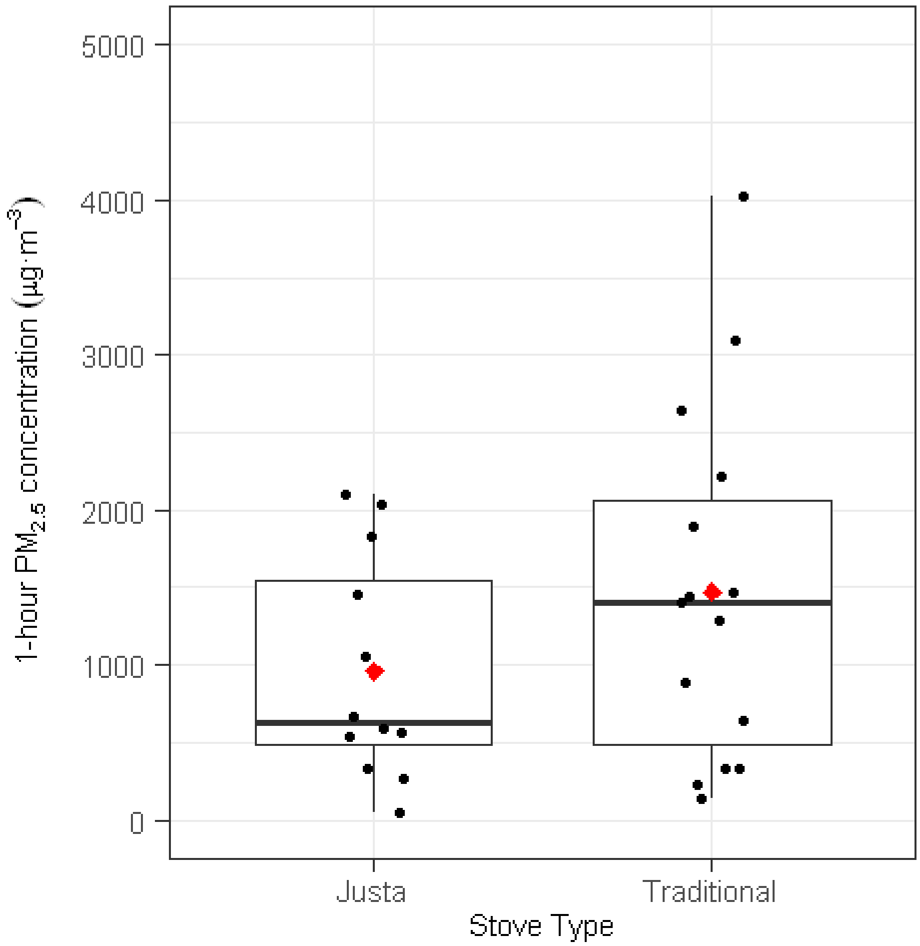

Figure 3: Maximum 1-hour average $PM_{2.5}$ concentrations measured in kitchens using traditional and cleaner-burning Justa stoves (N = 27) in rural Honduras. Black dots represent the observed concentrations. The lower boundary of the box represents the 25th percentile; the line within the box is the median; the upper boundary represents the 75th percentile. Bars indicate the 10th and 90th percentiles and the *"◊" represents the mean.*

Table 3: Number of Hours Spent Above 100 μg/m^3							
	Mean	Standard Deviation	Min	25th Percentile	Median	75th Percentile	Max
All samples (n=27)	4.0	3.7	0.43	1.6	2.3	5.2	15.2
Justa (n=12)	2.3	1.3	0.43	1.4	2.1	3.1	4.7
Traditional (n= 15)	5.5	4.4	1.1	1.8	4.7	8.3	15.6

*Corrected to gravimetric samples

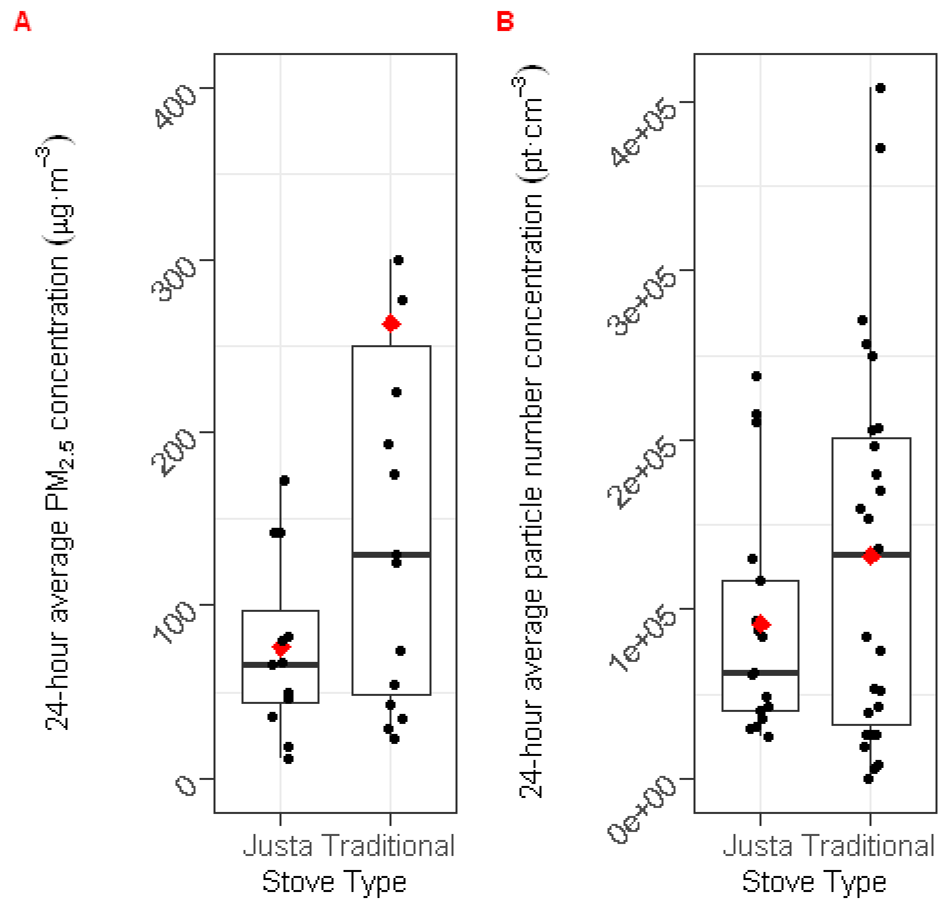

Figure 4: A: 24-hour average $PM_{2.5}$ (n=27), B: 24-hour particle number concentration (n=44). Measurements were in kitchens using traditional and cleaner-burning Justa cookstoves in rural Honduras. Dots represent the observed concentrations. The lower boundary of the box represents the 25th percentile; the line within the box is the median; the upper boundary represents the 75th percentile. Bars indicate the 10th and 90th percentiles and the "◊" represents the mean.

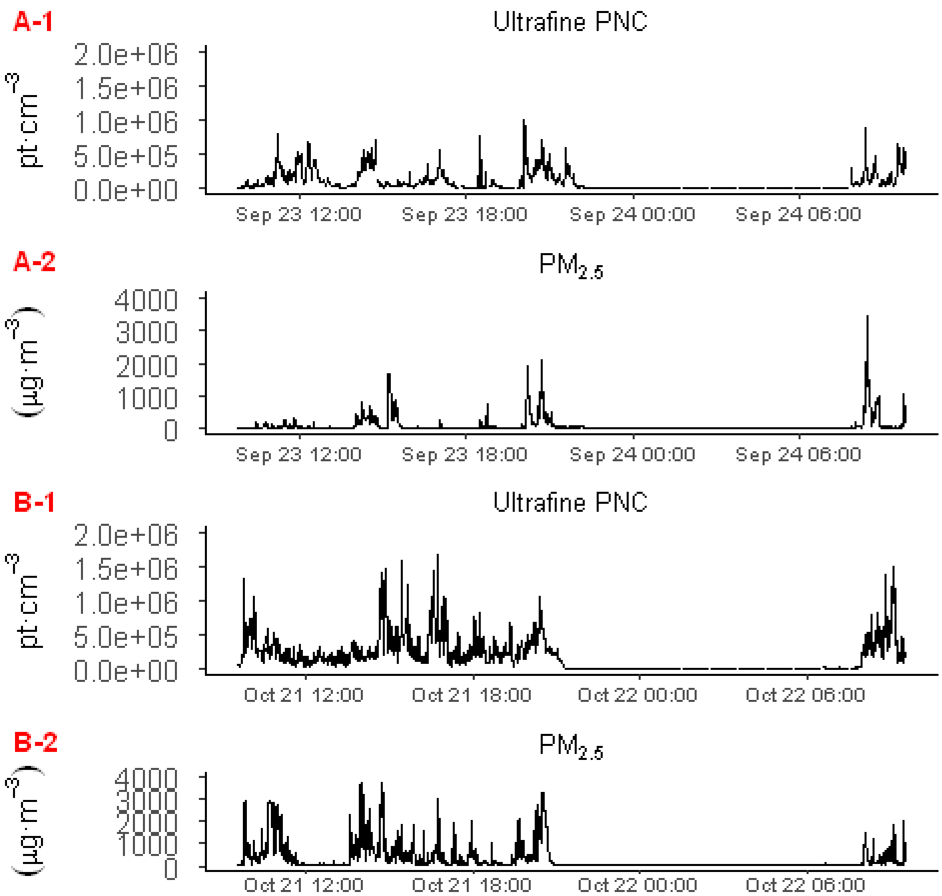

Figure 5: Example of real-time minute-level kitchen concentrations of PNC and $PM_{2.5}$ mass over a 24-hour time period. (A-1: Justa stove PNC, A-2: Justa stove $PM_{2.5}$, B-1: Traditional stove PNC, B-2: Traditional stove $PM_{2.5}$)

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