Published in final edited form as:
Environ Pollut. 2019 July ; 250: 251–261. doi:10.1016/j.envpol.2019.03.121.

VARIATION IN GRAVIMETRIC CORRECTION FACTORS FOR NEPHELOMETER-DERIVED ESTIMATES OF PERSONAL EXPOSURE TO $PM_{2.5}$

Jessica Tryner[a], **Nicholas Good**[b], **Ander Wilson**[c], **Maggie L. Clark**[b], **Jennifer L. Peel**[b], and **John Volckens**[a,*]

[a]Department of Mechanical Engineering, Colorado State University, 1374 Campus Delivery, Fort Collins, Colorado, United States 80523

[b]Department of Environmental and Radiological Health Sciences, Colorado State University, F1681 Campus Delivery, Fort Collins, Colorado, United States 80523

[c]Department of Statistics, Colorado State University, 1877 Campus Delivery, Fort Collins, Colorado, United States 80523

Abstract

Many portable monitors for quantifying mass concentrations of particulate matter air pollution rely on aerosol light scattering as the measurement method; however, the relationship between scattered light (what is measured) and aerosol mass concentration (the metric of interest) is a complex function of the refractive index, size distribution, and shape of the particles. In this study, we compared 33-hour personal $PM_{2.5}$ concentrations measured simultaneously using nephelometry (personal DataRAM pDR-1200) and gravimetric filter sampling for working adults (44 participants, 249 samples). Nephelometer- and filter-derived 33-hour average $PM_{2.5}$ concentrations were correlated (Spearman's $\rho = 0.77$); however, the nephelometer-derived concentration was within 20% of the filter-derived concentration for only 13% of samples. The nephelometer/filter ratio, which is used to correct light-scattering measurements to a gravimetric sample, had a median value of 0.52 and varied by over a factor of three (10^{th} percentile = 0.35, 90^{th} percentile = 1.1). When 33-hour samples with >50% of 10-s average nephelometer readings below the nephelometer limit of detection were removed from the dataset during sensitivity analyses, the fraction of nephelometer-derived concentrations that were within 20% of the filter-derived concentration increased to 25%. We also evaluated how much the accuracy of nephelometer-derived concentrations improved after applying: (1) a median correction factor derived from a subset of 44 gravimetric samples, (2) participant-specific correction factors derived from one same from each subject, and (3) correction factors predicted using linear models based on other variables recorded during the study. Each approach independently increased the fraction of nephelometer-derived concentrations that were within 20% of the filter-derived concentration to

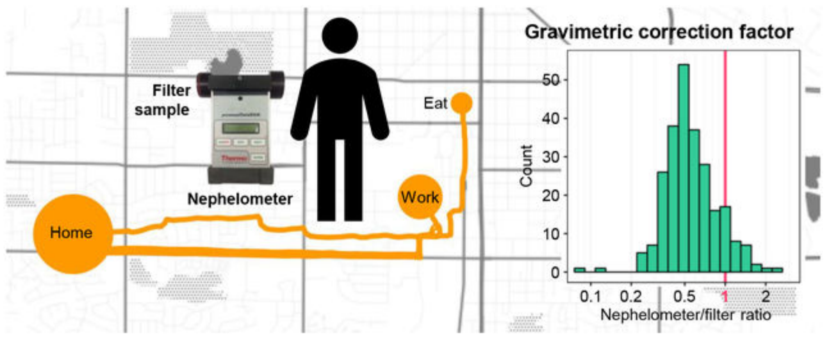
*Corresponding author: john.volckens@colostate.edu.

approximately 45%. These results illustrate the challenges with using light scattering (without correction to a concurrent gravimetric sample) to estimate personal exposure to $PM_{2.5}$ mass among mobile adults exposed to low daily average concentrations (median = 8 $\mu g \cdot m^{-3}$ in this study).

CAPSULE:

Variations in the factor used to correct nephelometer data to a gravimetric sample present challenges for estimating personal exposure to $PM_{2.5}$ mass.

Graphical Abstract



INTRODUCTION

Many studies rely on devices that measure light scattering to estimate concentrations of particulate matter (PM) air pollution (Howard-Reed et al., 2000; Lanki et al., 2002; Liu et al., 2002; Allen et al., 2004; Wallace et al., 2006; Chowdhury et al., 2007; Fischer and Koshland, 2007; Wallace et al., 2011; Gao et al., 2015; Pokhrel et al., 2015; Steinle et al., 2015; Kelly et al., 2017; Patel et al., 2017). Compared to filter-based methods, light-scattering sensors can be more affordable, more portable, and provide time-resolved estimates of PM mass concentration. As a result, light-scattering devices can increase the spatial and temporal resolution of measurements beyond what is typically possible with filter-based technologies. Such increases in measurement resolution can help identify the locations, activities, and pollution sources that contribute the most to exposure, thereby informing policies and behavioral changes designed to reduce exposures (Adams et al., 2009; Howard-Reed et al., 2000).

Epidemiological studies have linked exposure to higher PM mass concentrations with adverse health outcomes and mass-based air quality standards are used around the world (Brunekreef and Holgate, 2002; Pope and Dockery, 2006). As a result, the aerosol metric of interest is typically the mass concentration of PM to which an individual is exposed; however, light-scattering devices do not measure PM mass directly. The relationship between the amount of light scattered by an aerosol and the mass concentration of that aerosol is dependent on the refractive index, size, shape, number concentration, and density of the particles. For particles with diameters between 0.05 and 100 μm, this relationship is governed by complex Mie theory (Hinds, 1999). Because detailed particle property data are typically unavailable at the time and place of measurement, the accuracy of PM mass concentrations reported by light-scattering devices is uncertain.

Previous studies reported that measurements recorded by wearable, research-grade nephelometers (i.e., various models of the personal DataRAM) were sensitive to relative humidity (RH) and the type of aerosol sampled (Benton-Vitz and Volckens, 2008; Chakrabarti et al., 2004; Fischer and Koshland, 2007; Jenkins et al., 2004; Quintana et al., 2000; Singer and Delp, 2018; Sioutas et al., 2000; Sousan et al., 2017; Zhang et al., 2018). Multiple studies reported that, in accordance with Mie theory (Hinds, 1999), the nephelometer response was dependent upon the mass median diameter (MMD) of the sampled aerosol (Chakrabarti et al., 2004; Sioutas et al., 2000; Zhang et al., 2018). In addition, studies have reported that readings from low-cost light-scattering sensors—which have become popular in recent years and rely on the same physical principles as research-grade nephelometers to estimate PM concentrations—are sensitive to variations in particle refractive index, size, shape, and density (Wang et al., 2015; Austin et al., 2015; Liu et al., 2017; Sousan et al., 2017; Singer and Delp, 2018).

In personal monitoring applications, r^2 values ranging from 0.48 to 0.86 were reported for 24-hour average PM concentrations measured using personal DataRAM nephelometers and gravimetric samplers (Allen et al., 2004; Howard-Reed et al., 2000; Lanki et al., 2002; Liu et al., 2002; Wallace et al., 2006, 2011). Few studies have evaluated low-cost light-scattering devices for use in personal exposure monitoring (Steinle et al., 2015), which may prove more challenging than stationary monitoring due to the wider range of motion, environmental conditions, and aerosols to which the sensors could be exposed.

Research-grade nephelometers, like the personal DataRAM, often include a filter cartridge downstream of the light-scattering sensor. Using this arrangement, the PM concentration reported by the sensor can be corrected to a gravimetric sample in post-processing. The nephelometer/filter ratio, defined herein as the ratio of the time-averaged $PM_{2.5}$ concentration derived from nephelometer measurements to the time-averaged $PM_{2.5}$ concentration derived from a filter sample (both in $\mu g \cdot m^{-3}$), is the factor used to correct the time-resolved light-scattering measurement to the gravimetric sample.

Although instruments that measure the particle properties that influence the gravimetric correction factor (*e.g.*, composition, size distribution, and shape) are not amenable to personal sampling (due to cost and size), many parameters that might be correlated with particle properties (*e.g.*, black carbon concentration, particle number concentration, temperature, GPS location of the subject) can be practically measured during personal sampling campaigns (Adams et al., 2009; Good et al., 2016). As a result, one might be able to predict the correction factor (in the absence of a gravimetric sample) using these other variables.

In this study, the mass of $PM_{2.5}$ measured by a wearable aerosol nephelometer was compared to integrated filter measurements collected simultaneously during personal monitoring of working adults exposed to relatively low daily average $PM_{2.5}$ mass concentrations. We aimed to answer the following questions: (1) How well do personal

$PM_{2.5}$ mass concentrations measured using a nephelometer agree with personal $PM_{2.5}$ mass concentrations measured using a gravimetric sample (*i.e.*, are the two measures correlated and how accurate are the light-scattering measurements relative to the gravimetric measurements); (2) how much does the nephelometer/filter ratio vary during personal exposure assessment; (3) how much do constant or participant-specific correction factors derived from a subset (< 20%) of gravimetric samples improve the accuracy of nephelometer-derived concentrations; and (4) how much do correction factors predicted using other variables recorded in this personal exposure study improve the accuracy of nephelometer-derived concentrations? The consistency and predictability of the ratio used to correct nephelometer data to gravimetric measurements have important implications for the accuracy of measurements collected using light-scattering sensors in the absence of collocated gravimetric samples.

METHODS

Data Collection

Data were collected as part of the Fort Collins Commuter Study (Good et al., 2016). Study participants were working adults (18 to 65 years old) who lived in the Fort Collins area and commuted at least 1.5 miles from home to work. Participants also had to possess a valid driver's license, be a non-smoker, and have no regular occupational exposure to dust or fumes (Good et al., 2016). Participants carried a backpack (see Figure S1) containing equipment that measured their location and personal exposure to air pollution ($PM_{2.5}$ mass, $PM_{2.5}$ black carbon, particle number concentration, and carbon monoxide) for approximately 33 hours while they went about their normal daily routine (which included commuting to and from work either by car or bike). Monitoring began around 3 pm on the day before the commutes (the "sample date") and ended around 12 am on the day after the commutes. Data were collected between September 2012 and February 2014. Each participant was scheduled to complete eight monitoring periods during a 4- to 12-week period. The study continued throughout all seasons, but efforts were made to collect all samples for a given participant within a single season. In total, 377 samples were collected by 45 participants on 107 unique sample dates.

Each participant was randomly assigned a backpack full of sampling equipment on each sample date. The sampling equipment in the backpack included a pDR-1200 nephelometer (Thermo Fisher Scientific, Waltham, MA, USA), which featured a $PM_{2.5}$ inlet (PEM, SKC, Eighty Four, PA, USA) and a 37-mm filter on the outlet (PallFlex Fiberfilm T60A20; Pall, Port Washington, NY, USA). The pDR-1200 used an LED light source with a center wavelength of 880 nm, detected scattered light over an angle of 50° to 90°, and recorded data continuously using a 10-second averaging window. Airflow through the $PM_{2.5}$ inlet was maintained at 4.0 $L{\cdot}min^{-1}$ using a personal sampling pump (OMNI 400, Mesa Labs, Lakewood, CO, USA). Airflow through the nephelometer and filter was maintained at 3.8 $L{\cdot}min^{-1}$, while airflow through an aethalometer (microAeth Model AE51, AethLabs, San Francisco, CA, USA) installed in parallel with the nephelometer/filter was maintained at 0.2 $L{\cdot}min^{-1}$. To determine the mass of fine particulate black carbon to which the participant was exposed during the sample, aethalometer black carbon measurements were analyzed and integrated as described previously (Good et al., 2017). The black carbon fraction of $PM_{2.5}$ was calculated using the mass accumulated on the filter as the reference.

Other data collected included particle number concentration (for 128/377 samples; DiSC Mini, Matter Aerosol AG, Wohlen, Switzerland); carbon monoxide mixing ratio (T15n, Langan Products, San Francisco, CA, USA); GPS location and movement of the backpack (BT-Q1000XT, QStarz, Taipei, Taiwan); movement and heart rate of the participant (Actiheart, CamNtech, Cambridge, UK); as well as the temperature, RH, and light intensity measured on the outside of the backpack (MSR Electronics, GmbH, Seuzach, Switzerland). The procedures used to collect these data have been described previously (Good et al., 2016). Location data (GPS) and time-activity diaries were used to determine the amount of time that each participant spent in five microenvironment categories: home, work, transit, eatery, and other. The temperature, RH, and light intensity data (as measured on the backpack) were used to determine a change in microenvironment more precisely. Activity data were combined with time-resolved $PM_{2.5}$ measurements (taken by the pDR-1200) to estimate the fraction of the total $PM_{2.5}$ exposure associated with each microenvironment category.

Hourly ambient $PM_{2.5}$ concentrations in Fort Collins during the study period (September 2012–February 2014), as measured at EPA monitoring site 08-069-0009 using a ThermoFisher Scientific 1405-DF TEOM, were downloaded from the EPA AQS Data Mart (US EPA, 2015). Hourly ambient concentrations were averaged over the ~33-hour period associated with each sample for comparison to the average personal $PM_{2.5}$ concentration measured using the filter behind the nephelometer. Ambient temperature and RH data were obtained from the Christman Field weather station at Colorado State University ("Christman Field Data Access," 2017).

Quality Assurance

Prior to study initiation, all six pDR-1200 units were sent to the manufacturer for calibration. The factory calibration aerosol was Society of Automotive Engineers (SAE) Fine test dust, which has an MMD of 2–3 μm, a geometric standard deviation (GSD) of 2.5, a particle density of 2.60–2.65 $g \cdot cm^{-3}$, and a refractive index of 1.54 (Thermo Fisher Scientific, 2013). Prior to the start of each sample, each pDR was flushed with HEPA-filtered air and zeroed. Additionally, the light-scattering chamber was opened, cleaned with compressed air, and re-sealed once per month. Flow rates were checked at the inlet to the pDR and the inlet to the sample pump, at the start and end of each sampling period, using a BGI TriCal (Mesa Labs, Butler, NJ, USA).

Filters were pre- and post-weighed on a balance with 1 μg resolution (MX5, Mettler-Toledo, Columbus, OH, USA). The calibration of the balance was checked each day using a 50 mg calibration weight. Filters were equilibrated in the low-humidity, climate-controlled microbalance laboratory for at least 24 hours before weighing. Pre- and post-sample filter masses were measured in duplicate; if the first and second measurements differed by more than 5 μg, a third measurement was taken. Immediately prior to each measurement, the filter was placed on a Polonium-210 neutralizer (Staticmaster 2U500, NRD Static Control, Grand Island, NY, USA) for 10 seconds to eliminate static charge. The duplicate or triplicate measurements were averaged to obtain the pre- and post-sample filter weights used in all calculations.

The limit of detection (LOD) for the filter samples was calculated from the change in mass of 203 blanks collected over the duration of the study (one or two per sample date). Blank filters were pre-weighed, loaded into filter cartridges, removed from their filter cartridges, and post-weighed using the same procedures applied to the sample filters; however, the blank filters never left the laboratory. The filter LOD was calculated as three times the standard deviation of the mass accumulated on the blanks ($3s_{blank}$) and was equal to 31 μg (which corresponded to a 33-hour average concentration of 4 $\mu g \cdot m^{-3}$). The LOD of the pDR-1200 was estimated to be 3 $\mu g \cdot m^{-3}$ based on experiments conducted in a laboratory aerosol chamber (Good et al., 2016). Ten-second average $PM_{2.5}$ concentrations recorded by the nephelometer that were below the LOD were replaced with $3/\sqrt{2}\mu g \cdot m^{-3}$. For the 333/377 samples for which nephelometer data were available, the fraction of 10-s concentrations in a given 33-hour sample that were below the LOD ranged from 0% to 98% with a median of 60%.

The RH measured on the backpack was used to calculate dry $PM_{2.5}$ mass ($PM_{2.5,dry}$; $\mu g \cdot m^{-3}$) from each LOD-adjusted 10-s average data point recorded by the nephelometer ($PM_{2.5,wet}$; $\mu g \cdot m^{-3}$) as shown in Equation 1 (Chakrabarti et al., 2004). This RH correction was selected because Chakrabarti et al. (2004) reported that the relationship between $PM_{2.5,wet}$ and $PM_{2.5,dry}$ given by Equation 1 agreed with measurements taken by pDR-1200 nephelometers sampling ambient air at a stationary location in Los Angeles.

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

If the RH sensor on the backpack malfunctioned during a sample (n = 71/333), RH was assumed to be 30%. This assumed RH resulted in essentially no adjustment being applied to the nephelometer data using Equation 1. This assumption was considered reasonable because the 33-hour average RH measured on the outside of the backpack was 30% ± 10% for 60% of the samples for which the RH sensor did not malfunction. As a result of applying Equation 1, the 33-hour average nephelometer-derived $PM_{2.5}$ concentration decreased by a median value of 6% (see Figure S2).

The LOD- and RH-corrected 10-second average $PM_{2.5}$ concentrations derived from the nephelometer measurements were averaged over the entire sample period (~33 hours) for comparison to the average $PM_{2.5}$ concentration derived from the filter sample. Samples were only included in the main analysis if: (1) 10-second average nephelometer measurements were available for at least 85% of the sample period (309/377), (2) the mass accumulated on the filter was above the LOD (292/377 samples), and (3) the filter-derived $PM_{2.5}$ concentration was less than 145 $\mu g \cdot m^{-3}$ (375/377). A total of 249 samples, collected by 44 participants on 100 sample dates, were retained after applying these three criteria. The two filter-derived concentrations greater than 145 $\mu g \cdot m^{-3}$ were suspected to be erroneous, since they were: (a) over an order of magnitude higher than the median filter-derived concentration (7 $\mu g \cdot m^{-3}$ for all 377 samples), (b) more than 50% higher than the next highest filter-derived concentration (90 $\mu g \cdot m^{-3}$), and (c) not corroborated by higher-than-average nephelometer-derived concentrations.

Data Analyses

The first objective was to assess how well the $PM_{2.5}$ mass concentrations measured using the nephelometer agreed with the $PM_{2.5}$ mass concentrations measured using the filter sample. Spearman's rho (ρ) was calculated to evaluate rank-order correlation between the 33-hour average nephelometer-derived concentration and the filter-derived concentration. Spearman's rho is a measure of a monotonic relationship between two variables—the relationship need not be linear (Reimann et al., 2008). To evaluate the accuracy of the nephelometer measurements, absolute and percent differences between the 33-hour average nephelometer- and filter-derived $PM_{2.5}$ mass concentrations were calculated.

The second objective was to determine how much the nephelometer/filter ratio varied. The nephelometer/filter ratio is the factor that would be used to correct the light-scattering measurements to the gravimetric sample in post-processing (Thermo Fisher Scientific, 2013). For each sample, we calculated this ratio as the 33-hour average $PM_{2.5}$ concentration derived from the nephelometer measurements divided by the 33-hour average $PM_{2.5}$ concentration derived from the filter sample. To assess the fraction of the variance in the nephelometer/filter ratio that could be explained by (1) differences between versus within participants and (2) differences between versus within sample dates, we fitted separate one-way random effects models and estimated intraclass correlation coefficients (ICC) (see Supporting Information Section S1.1) (McGraw and Wong, 1996; Neter and Wasserman, 1974). When calculating the ICC, the nephelometer/filter ratio was log-transformed to satisfy model assumptions.

Sensitivity Analyses

Sensitivity analyses were conducted to examine how the results of the analyses described above were affected by: (a) not replacing 10-second average nephelometer readings below the LOD with $3/\sqrt{2}$ and (b) filtering the data based on the number of 10-s nephelometer readings equal to zero or below the LOD. For the data used in the sensitivity analyses, raw 10-second average concentrations recorded by the nephelometer were not adjusted based on the LOD of the instrument. Instead, all raw 10-second values (regardless of concentration) were RH-corrected and then averaged over the 33-hour sample period. The full dataset (377 samples) was then filtered in four steps based on progressively more stringent criteria (see SI S1.2).

In the main analysis, and in each of the four filtering steps, samples were only included if 10-s average nephelometer measurements were available for at least 85% of the sample period (309/377 samples). The sensitivity of the results to this criterion was also investigated by considering data sets that only included samples for which 10-s average nephelometer measurements were available for at least 90% (294/377) and 95% (240/377) of the sample period (see SI S2.2).

Correcting Nephelometer-Derived Concentrations using a Subset of Gravimetric Samples

The third objective was to evaluate how much correction factors derived from a subset of gravimetric samples improved the accuracy of the nephelometer-derived concentrations. We evaluated two approaches that could reduce the number of filter samples that would need to be collected during a study. In both approaches, gravimetric correction factors were predicted using a subset of 44 samples; this number was selected because it was equal to the number of participants. The first approach was to obtain gravimetric correction factors for a random subset of 44 samples, and then correct all of the nephelometer-derived concentrations (n = 249) using the median factor calculated for that subset. The second approach was to obtain a gravimetric correction factor for the first sample collected by each participant, and then correct all samples collected by that participant (including the first) using that initial factor. The first approach adjusted for population effects, whereas the second approach adjusted for time-invariant subject-specific effects. The extent to which these two approaches improved the accuracy of the nephelometer-derived concentrations was assessed by comparing the filter- and nephelometer-derived $PM_{2.5}$ concentrations, before and after correction, using the following metrics: (1) the fraction of samples for which the absolute difference was ≤ 5 $\mu g \cdot m^{-3}$, (2) the fraction of samples for which the percent difference was ≤ 20%, (3) the median absolute difference, and (4) the median percent difference.

Prediction of the Gravimetric Correction Factor

The fourth objective was to evaluate how much model-predicted correction factors improved the accuracy of the nephelometer-derived concentrations. We developed linear mixed models to predict the gravimetric correction factor (*i.e.*, the nephelometer/filter ratio) using other variables recorded during the study. For this approach, K-fold cross-validation was employed (Hastie et al., 2009). The 44 participants represented in the main analysis were divided into $K = 5$ folds (four groups of 9 and one group of 8). For each fold, a training data set consisting of the samples in the other four folds was used to generate a linear mixed model with the log-transformed nephelometer/filter ratio as the response variable. The model generation process involved two steps, which were repeated for each of the five training data sets.

First, a series of 15 linear mixed models was developed—using the lme4 package in R (Bates et al., 2015)—with a single metric of interest as the fixed effect, a random participant intercept, and the logarithm of the nephelometer/filter ratio as the outcome variable. The 15 metrics considered were: fraction of $PM_{2.5}$ mass that was black carbon; time-averaged particle number concentration; time-averaged personal carbon monoxide mixing ratio; time-averaged ambient temperature in Fort Collins; participant age; the fraction of time spent at home, work, in transit, in an eatery, or elsewhere; and the fraction of $PM_{2.5}$ exposure received at home, work, in transit, in an eatery, or elsewhere. To make the fixed-effect coefficients comparable, all variables were standardized to have a mean of zero and unit variance. For each model, the 95% confidence interval for the fixed-effect coefficient was calculated using the lmerTest package (Kuznetsova et al., 2017).

Second, a single mixed model—including all fixed effects from the first step with 95% confidence intervals that did not include zero (with no interaction terms) and random participant intercept—was fit to the training data set (without standardizing the variables to have a mean of zero and unit variance). Because not all effects remained significant once combined into a single model, backward elimination of fixed effects was performed using the 'step()' function in the lmerTest package.

The fixed-effect coefficients and overall fixed intercepts from the five final mixed models (one for each of the five folds) were used to predict correction factors for each sample. For example, the model developed using a training set consisting of folds 2, 3, 4 and 5 was used to predict correction factors for samples in fold 1. This step was repeated five times to predict correction factors for the samples in all five folds. The extent to which the model-predicted correction factors improved the accuracy of the nephelometer-derived concentrations was assessed by comparing the filter- and nephelometer-derived $PM_{2.5}$ concentrations, before and after correction, using the aforementioned four metrics.

RESULTS AND DISCUSSION

All participants were adults who worked outside their homes, meaning that they transitioned between different microenvironments (*e.g.*, at home, in transit, at work) throughout the day. Example time-location data for four different participant-days are shown in Figure 1. Data on the distribution of participant age, gender, and time spent in five different microenvironment categories are shown in Table S1. On average, participants spent 58% of their time at home, 20% of their time at work, 6% of their time in transit, 2% of their time at an eatery, 8% of their time in another microenvironment, and nephelometer data were not available 6% of the time. A previous analysis of microenvironmental exposures indicated that participants were exposed to the lowest concentrations at work and the highest concentrations in eateries, but cumulative exposures were dominated by the home microenvironment (where participants spent most of their time) (Koehler et al., 2018).

Nephelometer data recorded during two example samples are shown in Figure 2. For both samples, the nephelometer detected expected variations in personal $PM_{2.5}$ mass concentration as a participant transitioned between microenvironments and activities. For the sample shown in the top panel, a peak in $PM_{2.5}$ exposure occurred shortly after 18:00 hours on the first day. This exposure, which was likely due to cooking (the participant indicated that they began frying food for dinner at 18:30), persisted through the remainder of the evening. Another, smaller, peak occurred around 6:30 the next morning, when the participant was making breakfast. The participant commuted to work shortly before 9:00 and then went to an eatery for lunch shortly before noon. A large peak in exposure occurred at the eatery. The participant then returned to work for the remainder of the afternoon before commuting back home at approximately 17:00. Exposures were lowest during the early morning, when the participant was at home and likely to be asleep, and during working hours. For this sample, the nephelometer/filter ratio was close to 1 and 95% of the 10-second average $PM_{2.5}$ concentrations recorded by the nephelometer were above the LOD. For most of the 249 samples retained in the main analysis, the nephelometer/filter ratio was less than one (median = 0.52), and a smaller fraction of 10-s average nephelometer readings were above the LOD (median = 38%).

A sample with a more typical nephelometer/filter ratio (0.55) and fraction of 10-s nephelometer readings above the LOD (45%) is shown in the bottom panel of Figure 2. This sample corresponds to the GPS data shown in gold in Figure 1. Despite the lower fraction of 10-s average nephelometer readings above the LOD, expected variations in personal exposure are evident. Most readings below the LOD were recorded during the early morning, when the participant was at home and likely to be asleep, and during working hours. A high-concentration exposure—likely due to aerosol emissions during cooking—occurred at 21:00 hours on the first day and persisted through the remainder of the evening. Exposures above the LOD were recorded each time the participant commuted from one location to another.

Nephelometer- and filter-derived 33-hour average personal $PM_{2.5}$ concentrations are compared in Table 1 for the main analysis and the sensitivity analyses. For the data set used in the main analysis, filter-derived 33-hour personal $PM_{2.5}$ concentrations ranged from 5 to 83 $\mu g{\cdot}m^{-3}$, with a median of 8 $\mu g{\cdot}m^{-3}$ (Figure S3). The nephelometer-derived 33-hour average personal $PM_{2.5}$ concentration was strongly correlated with the filter-derived concentration (Spearman's $\rho = 0.77$; Figure 3); however, the nephelometer tended to underestimate the filter-derived concentration. The absolute difference between the nephelometer- and filter-derived 33-hour average concentrations was ≤ 5 $\mu g{\cdot}m^{-3}$ for 73% of samples. A difference of 5 $\mu g{\cdot}m^{-3}$ is small from an absolute standpoint but represents a percent difference of 63% for the median concentration of 8 $\mu g{\cdot}m^{-3}$. The percent difference between the nephelometer- and filter-derived concentrations had a median value of 49% (Figure S5) and was less than 20% for only 32/249 samples.

The histogram of nephelometer/filter ratios shown in Figure 4 emphasizes that the nephelometer tended to underestimate the filter-derived $PM_{2.5}$ concentration. The nephelometer/filter ratio had a median value of 0.52 and was less than one for 88% of samples (Table 1). Contrary to this study, many previous studies reported that pDR nephelometers overestimated PM mass concentrations by a factor of approximately 1.5 relative to gravimetric measurements (Fischer and Koshland, 2007; Howard-Reed et al., 2000; Lanki et al., 2002; Liu et al., 2002; Wallace et al., 2006, 2011; Wu et al., 2005). This overestimation has been attributed to the high-density of the SAE Fine test dust used to calibrate the pDR (2.6 $g{\cdot}cm^{-3}$) as well as differences in particle size distribution and index of refraction between SAE Fine test dust and ambient aerosols (Howard-Reed et al., 2000; Liu et al., 2002; Molenar, n.d.; Wallace et al., 2006, 2011). In addition, size-selective inlets were not used in most of these studies (Fischer and Koshland, 2007; Howard-Reed et al., 2000; Liu et al., 2002; Wallace et al., 2006; Wu et al., 2005). The reason for the underestimation observed in the present study is unknown but might be related to the $PM_{2.5}$ inlet used on the pDR, the size distribution of the aerosols to which participants were exposed, and/or the low 33-hour average $PM_{2.5}$ concentrations to which participants were exposed. In laboratory studies, pDR nephelometers have been found to underestimate PM concentrations for some aerosol types (Benton-Vitz and Volckens, 2008; Jenkins et al., 2004; Sousan et al., 2017). Sioutas et al. (2000) and Zhang et al. (2018) reported that the aerosol mass median diameter (MMD) was the most important parameter affecting the pDR response. In both studies, the ratio of the pDR-reported concentration to the gravimetrically-determined concentration decreased as MMD decreased. Sioutas et al. (2000) reported that the nephelometer/gravimetric ratio was 0.7 for a MMD of 0.3 μm. Zhang et al. (2018) reported that the nephelometer/gravimetric ratio was ~0.5 for a MMD of ~0.2 μm. The MMD of the aerosols to which Fort Collins Commuter Study participants were exposed is unknown; however, individuals with occupational exposure to dust or fumes were not eligible to participate, and the $PM_{2.5}$ inlet would have prevented large particles from entering the nephelometer.

If the nephelometer measurements were accurate and precise, the histogram shown in Figure 4 would have a central value equal to approximately one and would span a narrow range (*e.g.*, 0.8 to 1.2 if all nephelometer-derived concentrations were within 20% of the filter-derived concentration). If the nephelometer measurements were inaccurate [as demonstrated in previous studies (Howard-Reed et al., 2000; Liu et al., 2002; Wallace et al., 2011; Lanki et al., 2002; Allen et al., 2004; Wallace et al., 2006)] but the nephelometer under- or overestimated the filter-derived concentration by a consistent factor, the histogram would have a central value that was not equal to one but would span a narrow range (*e.g.*, ± 20% around the median). The nephelometer/filter ratios have a median value of 0.52 and span a range that varies by more than a factor of 3 (10^{th}–90^{th} percentile = 0.35–1.1). These results illustrate that the nephelometer measurements were not accurate, nor was the factor that would be used to correct the nephelometer measurements to the gravimetric filter sample consistent between personal samples.

This analysis assumes that the filter-derived $PM_{2.5}$ concentration was the true exposure, and that disagreement between the nephelometer- and filter-derived 33-hour average concentrations was largely due to bias and imprecision in the nephelometer measurements; however, bias and imprecision in the filter measurements can also contribute to variability in the nephelometer/filter ratio. The filter concentrations were assumed to be unbiased because: (1) they were derived from direct measurements of $PM_{2.5}$ mass and (2) quality control procedures (*e.g.*, flow checks, collection of filter blanks) were implemented to detect possible sources of bias in the gravimetric measurements. Furthermore, comparisons of co-located filter samples from previous studies suggests that imprecision in gravimetric measurements would not account for the width of the histogram in Figure 4 (see SI Section S2.1) (Pillarisetti et al., 2019; Wendt, 2018).

One possible explanation for the wide range of nephelometer/filter ratios seen in Figure 4 is that each participant was exposed to different aerosols, with different compositions and size distributions, depending on where they lived and worked, and their daily activities. Nephelometer/filter ratios as a function of participant number are shown in Figure 5. The nephelometer/filter ratios for participant 32 varied by less than a factor two across eight sample dates; however, the nephelometer/filter ratios for participant 18 (n = 7) varied by a factor of eight. The intra-class correlation coefficient (ICC) calculated from a linear mixed model using repeated samples for each participant indicated that 27% of the variability of the log-transformed nephelometer/filter ratio was explained by differences between participants (Table 1) (Neter and Wasserman, 1974).

Another possible explanation for the range of nephelometer/filter ratios seen in Figure 4 is that exposures were affected by day-to-day variations in the concentration and properties of outdoor ambient aerosol. Nephelometer/filter ratios for samples collected by different participants on the same date are shown in Figure S7. On 2013/06/26, the nephelometer/filter ratios for four participants varied by a factor of less than 1.2; however, on 2013/02/11, the nephelometer/filter ratios for four participants varied by a factor of 5.4. The ICC calculated from a linear mixed model and repeated samples for each date indicated that differences between sample dates accounted 14% of the variance in the log-transformed ratio (Table 1).

Sensitivity Analyses

As filtering progressed from the 1st to 3rd steps in the sensitivity analyses, samples with high fractions of 10-second nephelometer readings equal to zero or below the nephelometer LOD (3 $\mu g \cdot m^{-3}$) were removed from the data set. In the 4th filtering step, samples that captured 33-hour average exposures below 4 $\mu g \cdot m^{-3}$ (as measured by the nephelometer) were removed. Between the first and fourth filtering steps, the median filter-derived personal $PM_{2.5}$ concentration increased from 8 to 12 $\mu g \cdot m^{-3}$ and the median fraction of 10-second nephelometer readings above the LOD increased from 38% to 72%. In addition, the median nephelometer/filter ratio increased from 0.44 to 0.74 and the fraction of nephelometer/filter ratios equal to 1.0 ± 0.2 increased from 11% to 29% (Figure S10). These results suggest that disagreement between the nephelometer- and filter-derived concentrations was exacerbated by the low concentrations to which participants were exposed and the large fraction of sub-LOD 10-second average nephelometer readings that were recorded during many samples as result.

Correcting Nephelometer-Derived Concentrations using a Subset of Gravimetric Samples

The inconsistency in the nephelometer/filter ratio shown in Figure 4 means that applying a single gravimetric correction factor to all of the nephelometer-derived concentrations would not result in accurate estimates of $PM_{2.5}$ exposure for all samples. When all 249 nephelometer-derived concentrations from the main analysis were corrected using the median gravimetric correction factor measured for a random subset of 44 samples (0.55), the median nephelometer/filter ratio shifted from 0.52 to 0.95 and the fraction of the nephelometer-derived concentrations that were within 20% of the filter-derived concentration increased from 13% to 43% (Table 2); however, the nephelometer/filter ratio still varied by a factor of 3 between the 10th and 90th percentiles.

Participant-specific correction factors did not perform better than the median correction factor calculated from a random subset of 44 samples. When the nephelometer-derived concentrations for each participant were corrected using the nephelometer/filter ratio measured during the first sample collected by each participant (see Figure 5), the median nephelometer/filter ratio shifted from 0.52 to 1.0 and the fraction of nephelometer-derived concentrations that were within 20% of the filter-derived concentration increased from 13% (32 samples) to 44% (109 samples); however, 44 of these 109 samples were adjusted using concurrently-measured gravimetric correction factors. Nephelometer-derived concentrations were within 20% of the filter-derived concentration for 65 samples that were corrected without concurrent filter samples (Table 2).

Estimates and 95% confidence intervals (CIs) for the fixed-effect coefficients associated with the 15 metrics tested as predictors in linear mixed models are shown in Figure S13 for each of the five training data sets. The final linear mixed models, which were used to predict correction factors for each fold, are shown in Equations 2–6. Equations 2–6 were obtained by fitting a single mixed model—including all fixed effects from Figure S13 with 95% confidence intervals that did not include zero (with no interaction terms) and random participant intercept—to each training data set and then performing backward elimination of fixed effects using the 'step()' function in the lmerTest package.

$$\log\left(\frac{PM_{2.5,\,neph}}{PM_{2.5,\,filter}}\right) = \alpha_i + \beta_1 f_{exp,\,transit} + \beta_2 \bar{T}_{ambient} + \beta_3 f_{exp,\,eat} + \varepsilon \quad (2)$$

$$\log\left(\frac{PM_{2.5,\,neph}}{PM_{2.5,\,filter}}\right) = \alpha_i + \beta_1 f_{exp,\,transit} + \beta_2 f_{exp,\,work} + \beta_3 f_{exp,\,eat} + \varepsilon \quad (3)$$

$$\log\left(\frac{PM_{2.5,\,neph}}{PM_{2.5,\,filter}}\right) = \alpha_i + \beta_1 f_{exp,\,transit} + \beta_2 f_{exp,\,work} + \beta_3 f_{time,\,home} + \varepsilon \quad (4)$$

$$\log\left(\frac{PM_{2.5,\,neph}}{PM_{2.5,\,filter}}\right) = \alpha_i + \beta_1 f_{exp,\,transit} + \beta_2 A + \varepsilon \quad (5)$$

$$\log\left(\frac{PM_{2.5,\,neph}}{PM_{2.5,\,filter}}\right) = \alpha_i + \beta_1 f_{exp,\,transit} + \beta_2 f_{exp,\,work} + \beta_3 f_{time,\,home} + \beta_4 f_{BC} + \varepsilon \quad (6)$$

where $PM_{2.5,neph}$ was the 33-hour average nephelometer-derived $PM_{2.5}$ concentration; $PM_{2.5,filter}$ was the filter-derived $PM_{2.5}$ concentration; α_i was the participant-specific random intercept, β_j was a fixed-effect coefficient; $f_{exp,transit}$, $f_{exp,eat}$, and $f_{exp,work}$ were the fractions of exposure received in transit, at an eatery, and at work, respectively; $f_{time,home}$ was the fraction of time spent at home; $\bar{T}_{ambient}$ was the mean ambient temperature in Fort Collins over the 33-hour period (°C); A was the participant age (years); f_{BC} was the fraction of $PM_{2.5}$ mass that was black carbon; and ε was the random error. Participant-specific intercepts were averaged when the models were used for prediction.

Four of the seven predictors that appeared in the final models were associated with specific microenvironments ($f_{exp,transit}$, $f_{exp,eat}$, $f_{exp,work}$, and $f_{time,home}$). The importance of these predictors might be explained by the variation in aerosol properties (and, consequently, the nephelometer response to a given mass concentration of aerosol) that would be expected between the different microenvironment categories (*e.g.*, in transit vs. at an eatery). The log-transformed nephelometer/filter ratio increased as the fraction of exposure received at an eatery increased. The log-transformed nephelometer/filter ratio decreased with increasing values of the fraction of exposure received in transit, the fraction of exposure received at work, and the fraction of time spent at home.

When the 33-hour average nephelometer-derived $PM_{2.5}$ concentrations were corrected using model-derived factors, the median nephelometer/filter ratio shifted from 0.52 to 0.98 and the fraction of the nephelometer-derived concentrations that were within 20% of the filter-derived concentration increased from 13% to 42% (Table 2).

CONCLUSIONS AND PRACTICAL IMPLICATIONS

The strong rank-order correlation between the nephelometer- and filter-derived concentrations ($\rho = 0.77$) indicates that portable light-scattering instruments can provide useful information about relative $PM_{2.5}$ concentrations. For example, Gao et al. (2015) reported that a network of low-cost light-scattering sensors could be used to detect a "hotspot" of ambient $PM_{2.5}$ pollution within a large city. Using the nephelometer data presented here, Good et al. (2016) were able to determine that adults who commuted to work by bicycle were exposed to higher $PM_{2.5}$ concentrations than adults who commuted to work by car. In other words, the nephelometer data were useful for examining relative differences in exposure within a single microenvironment category (in transit).

Without correction to gravimetric samples, however, research-grade nephelometers provided accurate (± 20%) estimates of absolute 33-hour average personal $PM_{2.5}$ concentrations for only 13% of samples collected by working adults exposed to low daily average concentrations (median = 8 $\mu g \cdot m^{-3}$). The results of the sensitivity analyses suggested that the accuracy of the nephelometer-derived 33-hour average concentrations was sensitive to the number of 10-s average nephelometer readings below the nephelometer limit of detection. These limitations of light-scattering instruments are important to keep in mind, especially when one wishes to evaluate absolute, as opposed to relative, personal exposures.

Figure 5 Nephelometer/filter ratios derived from repeated measurements (n = 249 samples) on 44 adults living in the same city varied by a factor of three between the 10^{th} percentile (0.35) and the 90^{th} percentile (1.1), meaning that the calibration factor used to correct the nephelometer measurements to gravimetrically-determined $PM_{2.5}$ concentrations did not remain constant. When nephelometer-derived concentrations were corrected using a median gravimetric correction factor calculated from a random subset of 44 samples, the fraction of the nephelometer-derived concentrations that were within 20% of the filter-derived concentration increased to 43%. This result indicates that collecting gravimetric measurements for just a subset of samples can improve the accuracy of nephelometer-derived estimates of personal exposure to $PM_{2.5}$; however, accuracy remained less than 20%

for the majority of samples. Neither constant participant-specific correction factors (calculated from the first gravimetric sample collected by each participant) nor correction factors predicted using a more complicated linear modeling approach performed better than the constant correction factor calculated from a random subset of samples.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

ACKNOWLEDGMENTS

The authors thank the members of the Fort Collins Commuter Study research team: Anna Mölter, Charis Ackerson, Annette Bachand, Taylor Carpenter, Kristen M. Fedak, Ashleigh Kayne, Kirsten Koehler, Brianna Moore, Christian L'Orange, Casey Quinn, Viney Ugave, and Amy L. Stuart.

Funding: This work was funded by the United States Department of Health and Human Services (HHS), National Institute of Health (NIH), National Institute of Environmental Health Sciences (NIEHS) under grant ES020017 and under grant OH009229 by CDC NIOSH Mountain and Plains Education and Research Center. The funding sources had no role in the study design; in collection, analysis, and interpretation of data; in the writing of this article; or in the decision to submit this article for publication.

REFERENCES

Adams C, Riggs P, Volckens J, 2009 Development of a method for personal, spatiotemporal exposure assessment. J. Environ. Monit 11, 1331–1339. 10.1039/b903841h [PubMed: 20449221]

Allen R, Wallace L, Larson T, Sheppard L, Liu L-JS, 2004 Estimated Hourly Personal Exposures to Ambient and Nonambient Particulate Matter Among Sensitive Populations in Seattle, Washington. J. Air Waste Manag. Assoc 54, 1197–1211. 10.1080/10473289.2004.10470988 [PubMed: 15468672]

Austin E, Novosselov I, Seto E, Yost MG, 2015 Laboratory Evaluation of the Shinyei PPD42NS Low-Cost Particulate Matter Sensor. PLOS ONE 10, e0137789 10.1371/journal.pone.0137789 [PubMed: 26367264]

Bates D, Mächler M, Bolker B, Walker S, 2015 Fitting Linear Mixed-Effects Models Using lme4. J. Stat. Softw 67 10.18637/jss.v067.i01

Benton-Vitz K, Volckens J, 2008 Evaluation of the pDR-1200 Real-Time Aerosol Monitor. J. Occup. Environ. Hyg 5, 353–359. 10.1080/15459620802009919 [PubMed: 18365888]

Brunekreef B, Holgate ST, 2002 Air pollution and health. The Lancet 360, 1233–1242. 10.1016/S0140-6736(02)11274-8

Chakrabarti B, Fine PM, Delfino R, Sioutas C, 2004 Performance evaluation of the active-flow personal DataRAM PM2.5 mass monitor (Thermo Anderson pDR-1200) designed for continuous personal exposure measurements. Atmos. Environ 38, 3329–3340. 10.1016/j.atmosenv.2004.03.007

Chowdhury Z, Edwards RD, Johnson M, Naumoff Shields K, Allen T, Canuz E, Smith KR, 2007 An inexpensive light-scattering particle monitor: field validation. J. Environ. Monit 9, 1099 10.1039/b709329m [PubMed: 17909644]

Christman Field Data Access [WWW Document], 2017 Colo. State Univ. Dep. Atmospheric Sci URL http://atmos.colostate.edu/fccwx/fccwx_data_form.php (accessed 10.3.17).

Fischer SL, Koshland CP, 2007 Field performance of a nephelometer in rural kitchens: effects of high humidity excursions and correlations to gravimetric analyses. J. Expo. Sci. Environ. Epidemiol 17, 141 10.1038/sj.jes.7500486 [PubMed: 16670712]

Gao M, Cao J, Seto E, 2015 A distributed network of low-cost continuous reading sensors to measure spatiotemporal variations of PM2.5 in Xi'an, China. Environ. Pollut 199, 56–65. 10.1016/j.envpol.2015.01.013 [PubMed: 25618367]

Good N, Mölter A, Ackerson C, Bachand A, Carpenter T, Clark ML, Fedak KM, Kayne A, Koehler K, Moore B, L'Orange C, Quinn C, Ugave V, Stuart AL, Peel JL, Volckens J, 2016 The Fort Collins

Commuter Study: Impact of route type and transport mode on personal exposure to multiple air pollutants. J. Expo. Sci. Environ. Epidemiol 26, 397–404. 10.1038/jes.2015.68 [PubMed: 26507004]

Good N, Mölter A, Peel JL, Volckens J, 2017 An accurate filter loading correction is essential for assessing personal exposure to black carbon using an Aethalometer. J. Expo. Sci. Environ. Epidemiol 27, 409–416. 10.1038/jes.2016.71 [PubMed: 28000686]

Hastie T, Tibshirani R, Friedman JH, 2009 The elements of statistical learning: data mining, inference, and prediction, 2nd ed. ed, Springer series in statistics. Springer, New York, NY.

Hinds WC, 1999 Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles, 2nd ed. John Wiley & Sons, New York.

Howard-Reed C, Rea AW, Zufall MJ, Burke JM, Williams RW, Suggs JC, Sheldon LS, Walsh D, Kwok R, 2000 Use of a Continuous Nephelometer to Measure Personal Exposure to Particles During the U.S. Environmental Protection Agency Baltimore and Fresno Panel Studies. J. Air Waste Manag. Assoc 50, 1125–1132. 10.1080/10473289.2000.10464150 [PubMed: 10939206]

Jenkins RA, Ilgner RH, Tomkins BA, Peters DW, 2004 Development and Application of Protocols for the Determination of Response of Real-Time Particle Monitors to Common Indoor Aerosols. J. Air Waste Manag. Assoc 54, 229–241. 10.1080/10473289.2004.10470892 [PubMed: 14977324]

Kelly KE, Whitaker J, Petty A, Widmer C, Dybwad A, Sleeth D, Martin R, Butterfield A, 2017 Ambient and laboratory evaluation of a low-cost particulate matter sensor. Environ. Pollut 221, 491–500. 10.1016/j.envpol.2016.12.039 [PubMed: 28012666]

Koehler K, Good N, Wilson A, Mölter A, Moore BF, Carpenter T, Peel JL, Volckens J, 2018 The Fort Collins Commuter Study: Variability in Personal Exposure to Air Pollutants by Microenvironment. Indoor Air 10.1111/ina.12533

Kuznetsova A, Brockhoff PB, Christensen RHB, 2017 lmerTest Package: Tests in Linear Mixed Effects Models. J. Stat. Softw 82 10.18637/jss.v082.i13

Lanki T, Alm S, Ruuskanen J, Janssen NAH, Jantunen M, Pekkanen J, 2002 Photometrically measured continuous personal PM2.5 exposure: Levels and correlation to a gravimetric method. J. Expo. Anal. Environ. Epidemiol 12, 172–178. 10.1038/sj.jea.7500218 [PubMed: 12032813]

Liu D, Zhang Q, Jiang J, Chen D-R, 2017 Performance calibration of low-cost and portable particular matter (PM) sensors. J. Aerosol Sci 112, 1–10. 10.1016/j.jaerosci.2017.05.011

Liu L-JS, Slaughter JC, Larson TV, 2002 Comparison of Light Scattering Devices and Impactors for Particulate Measurements in Indoor, Outdoor, and Personal Environments. Environ. Sci. Technol 36, 2977–2986. 10.1021/es0112644 [PubMed: 12144275]

McGraw KO, Wong SP, 1996 Forming inferences about some intraclass correlation coefficients. Psychol. Methods 1, 30–46. 10.1037/1082-989X.1.1.30

Molenar JV, n.d. Theoretical Analysis of $PM_{2.5}$ Mass Measurements by Nephelometry - #110. Air Resource Specialists, Inc., Fort Collins, CO.

Neter J, Wasserman W, 1974 Applied Linear Statistical Models: Regression, Analysis of Variance, and Experimental Designs Richard D. Irwin, Inc., Homewood, IL.

Patel S, Li J, Pandey A, Pervez S, Chakrabarty RK, Biswas P, 2017 Spatio-temporal measurement of indoor particulate matter concentrations using a wireless network of low-cost sensors in households using solid fuels. Environ. Res 152, 59–65. 10.1016/j.envres.2016.10.001 [PubMed: 27741449]

Pillarisetti A, Carter E, Rajkumar S, Young BN, Benka-Coker ML, Peel JL, Johnson M, Clark ML, 2019 Measuring personal exposure to fine particulate matter (PM2.5) among rural Honduran women: A field evaluation of the Ultrasonic Personal Aerosol Sampler (UPAS). Environ. Int 123, 50–53. 10.1016/j.envint.2018.11.014 [PubMed: 30496981]

Pokhrel AK, Bates MN, Acharya J, Valentiner-Branth P, Chandyo RK, Shrestha PS, Raut AK, Smith KR, 2015 PM 2.5 in household kitchens of Bhaktapur, Nepal, using four different cooking fuels. Atmos. Environ 113, 159–168. 10.1016/j.atmosenv.2015.04.060

Pope CA, Dockery DW, 2006 Health Effects of Fine Particulate Air Pollution: Lines that Connect. J. Air Waste Manag. Assoc 56, 709–742. 10.1080/10473289.2006.10464485 [PubMed: 16805397]

Quintana PJE, Samimi BS, Kleinman MT, Liu L-J, Soto K, Warner GY, Bufalino C, Valencia J, Francis D, Hovell MH, Delfino RJ, 2000 Evaluation of a real-time passive personal particle monitor in fixed site residential indoor and ambient measurements. J. Expo. Anal. Environ. Epidemiol 10, 437–445. [PubMed: 11051534]

Reimann C, Filzmoser P, Garrett RG, Dutter R, 2008 Statistical Data Analysis Explained: Applied Environmental Statistics with R John Wiley & Sons, Chichester, England.

Singer BC, Delp WW, 2018 Response of consumer and research grade indoor air quality monitors to residential sources of fine particles. Indoor Air 28, 624–639. 10.1111/ina.12463 [PubMed: 29683219]

Sioutas C, Kim S, Chang M, Terrell LL, Gong H, 2000 Field evaluation of a modified DataRAM MIE scattering monitor for real-time PM2.5 mass concentration measurements. Atmos. Environ 34, 4829–4838. 10.1016/S1352-2310(00)00244-2

Sousan S, Koehler K, Hallett L, Peters TM, 2017 Evaluation of consumer monitors to measure particulate matter. J. Aerosol Sci 107, 123–133. 10.1016/j.jaerosci.2017.02.013 [PubMed: 28871212]

Steinle S, Reis S, Sabel CE, Semple S, Twigg MM, Braban CF, Leeson SR, Heal MR, Harrison D, Lin C, Wu H, 2015 Personal exposure monitoring of PM 2.5 in indoor and outdoor microenvironments. Sci. Total Environ 508, 383–394. 10.1016/j.scitotenv.2014.12.003 [PubMed: 25497678]

Thermo Fisher Scientific, 2013 Model pDR-100AN/1200 personalDATARAM Instruction Manual Franklin, MA.

US EPA, 2015 AQS API / Query AirData [WWW Document] URL https://aqs.epa.gov/api (accessed 4.21.18).

Wallace L, Williams R, Rea A, Croghan C, 2006 Continuous weeklong measurements of personal exposures and indoor concentrations of fine particles for 37 health-impaired North Carolina residents for up to four seasons. Atmos. Environ 40, 399–414. 10.1016/j.atmosenv.2005.08.042

Wallace LA, Wheeler AJ, Kearney J, Van Ryswyk K, You H, Kulka RH, Rasmussen PE, Brook JR, Xu X, 2011 Validation of continuous particle monitors for personal, indoor, and outdoor exposures. J. Expo. Sci. Environ. Epidemiol 21, 49–64. [PubMed: 20502493]

Wang Y, Li J, Jing H, Zhang Q, Jiang J, Biswas P, 2015 Laboratory Evaluation and Calibration of Three Low-Cost Particle Sensors for Particulate Matter Measurement. Aerosol Sci. Technol 49, 1063–1077. 10.1080/02786826.2015.1100710

Wendt E, 2018 A low-cost monitor for simultaneous measurement of fine particulate matter and aerosol optical depth Colorado State University, Fort Collins, CO.

Wu C-F, Delfino RJ, Floro JN, Samimi BS, Quintana PJE, Kleinman MT, Liu L-JS, 2005 Evaluation and quality control of personal nephelometers in indoor, outdoor and personal environments. J. Expo. Anal. Environ. Epidemiol 15, 99–110. [PubMed: 15039794]

Zhang J, Marto JP, Schwab JJ, 2018 Exploring the applicability and limitations of selected optical scattering instruments for PM mass measurement. Atmospheric Meas. Tech 11, 2995–3005. 10.5194/amt-11-2995-2018

HIGHLIGHTS

- Nephelometer- and filter-derived $PM_{2.5}$ concentrations were correlated (ρ = 0.77)
- The nephelometer tended to underestimate the filter measurement by ~50%
- The nephelometer/filter ratio was sensitive to nephelometer readings below 3 $\mu g \cdot m^{-3}$
- Gravimetric correction factor varied by 300% between the 10^{th} and 90^{th} percentiles
- Modeled corrections brought 45% of nephelometer concentrations within 20% of filter

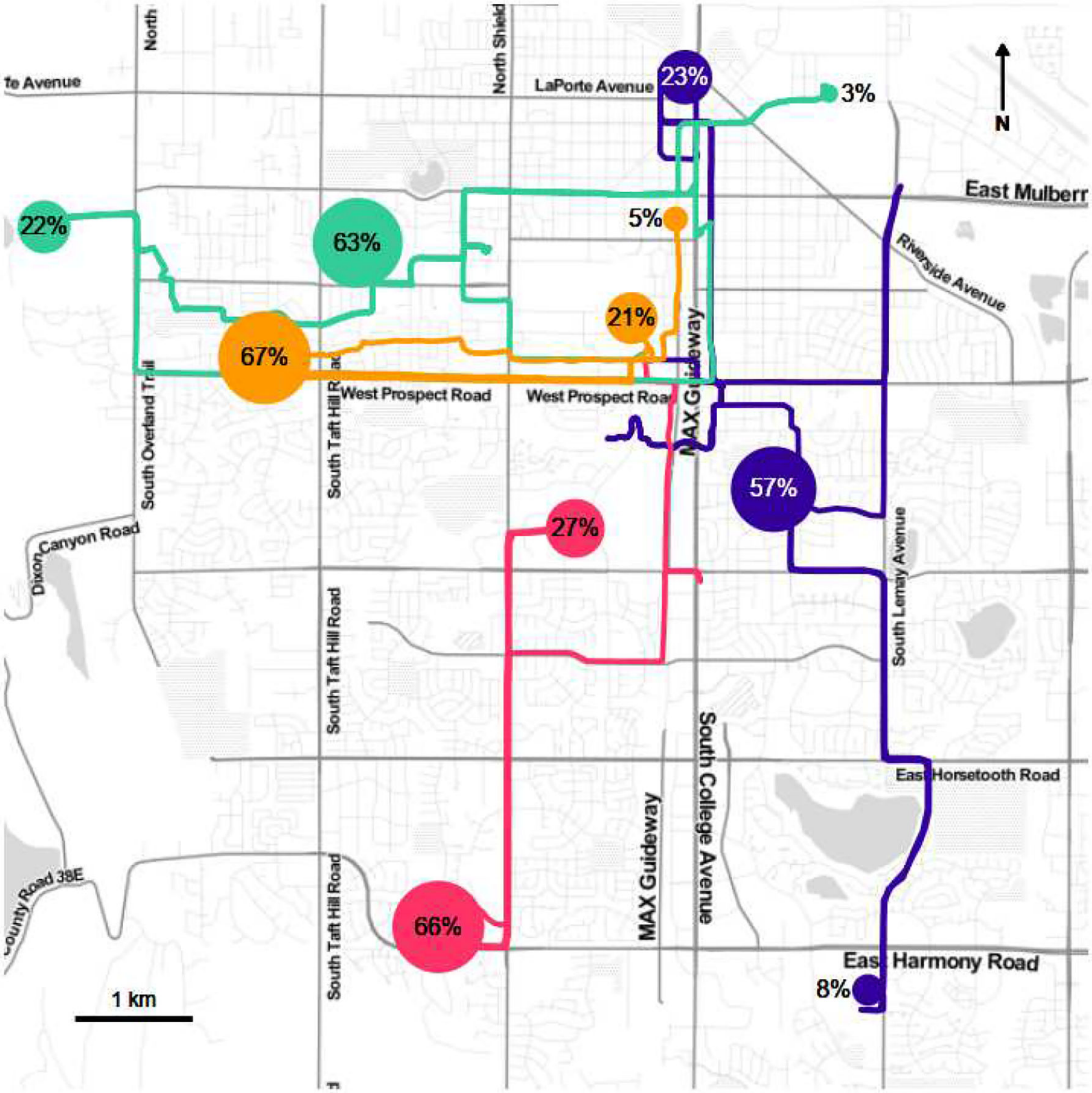


Figure 1.

GPS traces from single 33-hour samples collected by four participants. Circles represent time spent in stationary locations (*e.g.*, work, home, eatery). The percentage of time spent in each stationary location is noted, and the areas of the circles are proportional to these percentages. The percentage of time spent in transit varied from 3% to 8% for these participants.

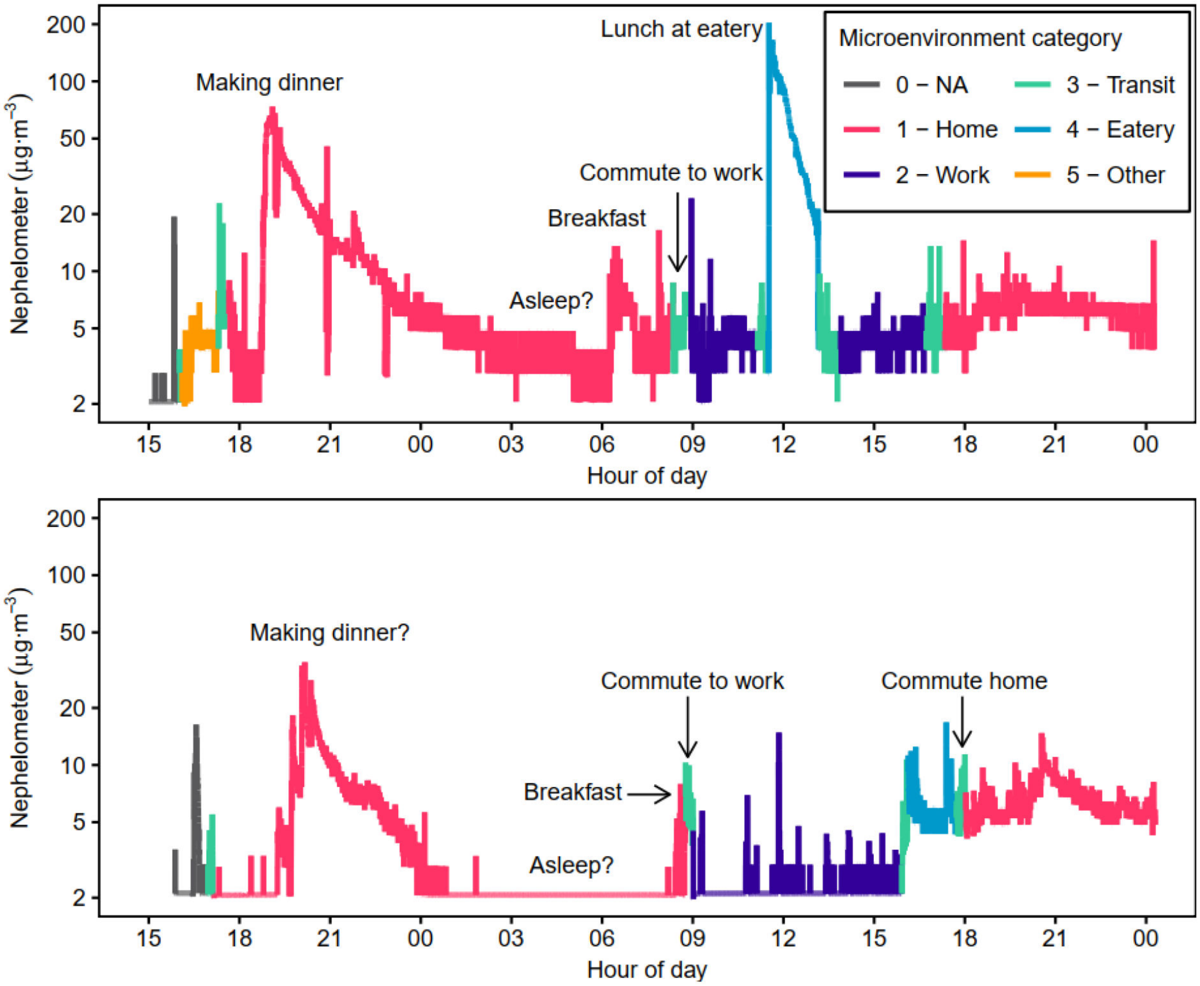


Figure 2.

The 10-second average LOD- and RH-corrected $PM_{2.5}$ concentrations recorded by the nephelometer during example 33-hour samples. Top: the filter-derived personal $PM_{2.5}$ concentration was 10 μg·m^{-3}, the nephelometer/filter ratio was 0.96, and 95% of the 10-second average nephelometer readings were above the limit of detection (LOD) of 3 μg/m^3. Bottom: the filter-derived personal $PM_{2.5}$ concentration was 8 μg·m^{-3}, the nephelometer/filter ratio was 0.55, and 45% of the 10-second nephelometer readings were above the LOD.

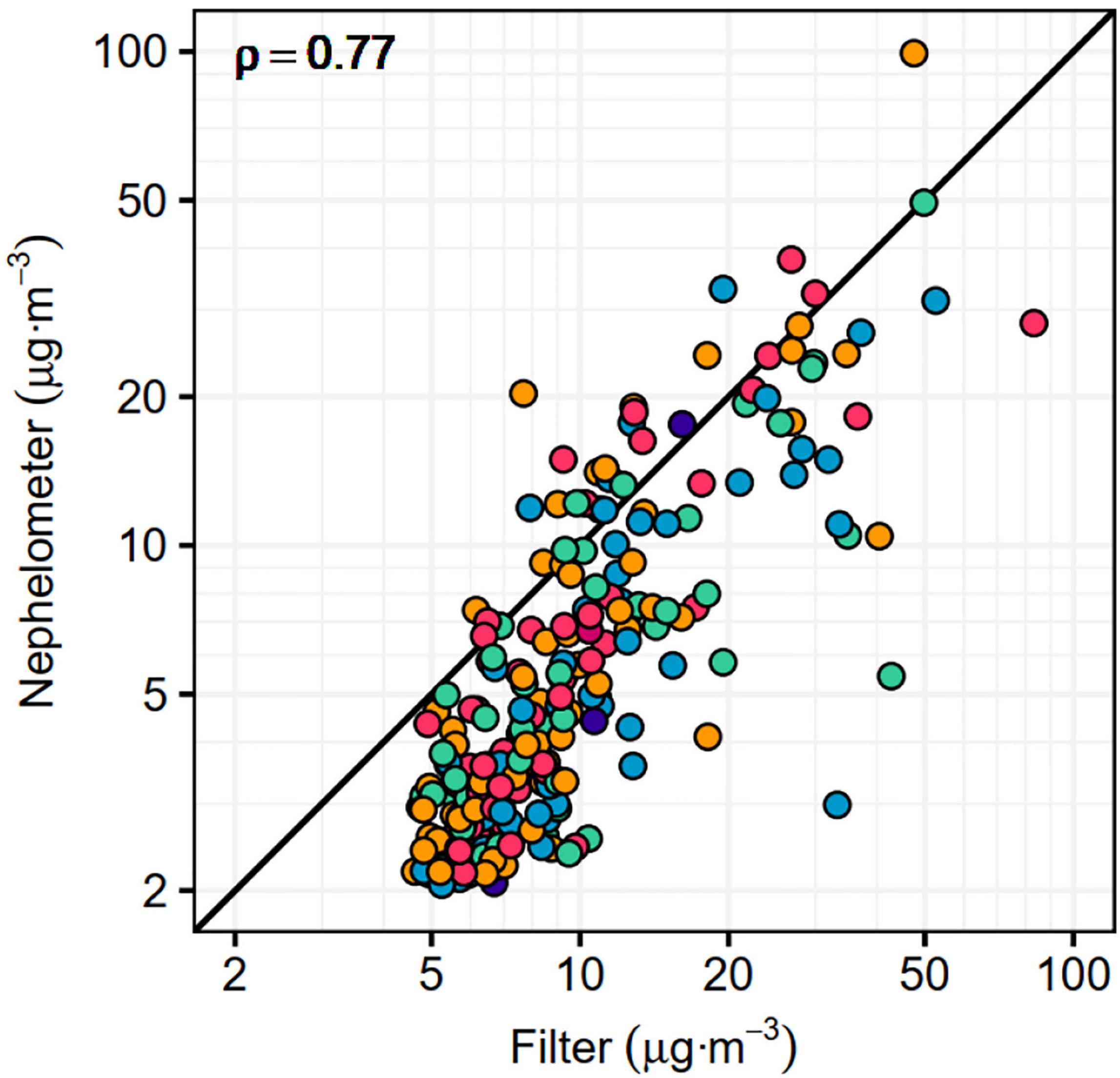


Figure 3.

A scatterplot of the 33-hour average $PM_{2.5}$ concentrations derived from the filter and nephelometer samples (n = 249). The solid line is y = x. Samples collected using different pDR-1200 units are shown in different colors (six total) to improve readability.

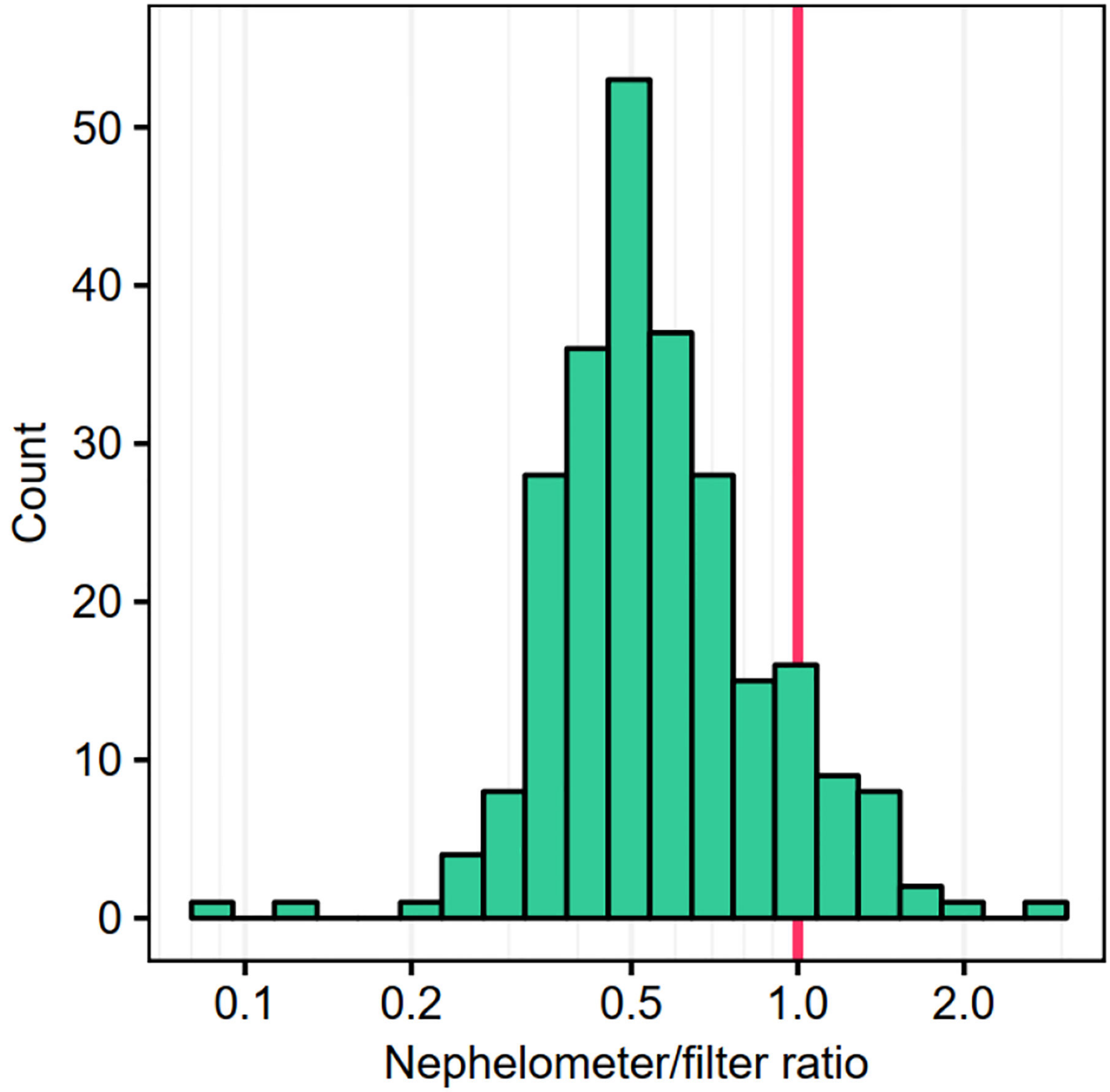


Figure 4.
A histogram of nephelometer/filter ratios for the data used in the main analysis (n = 249).

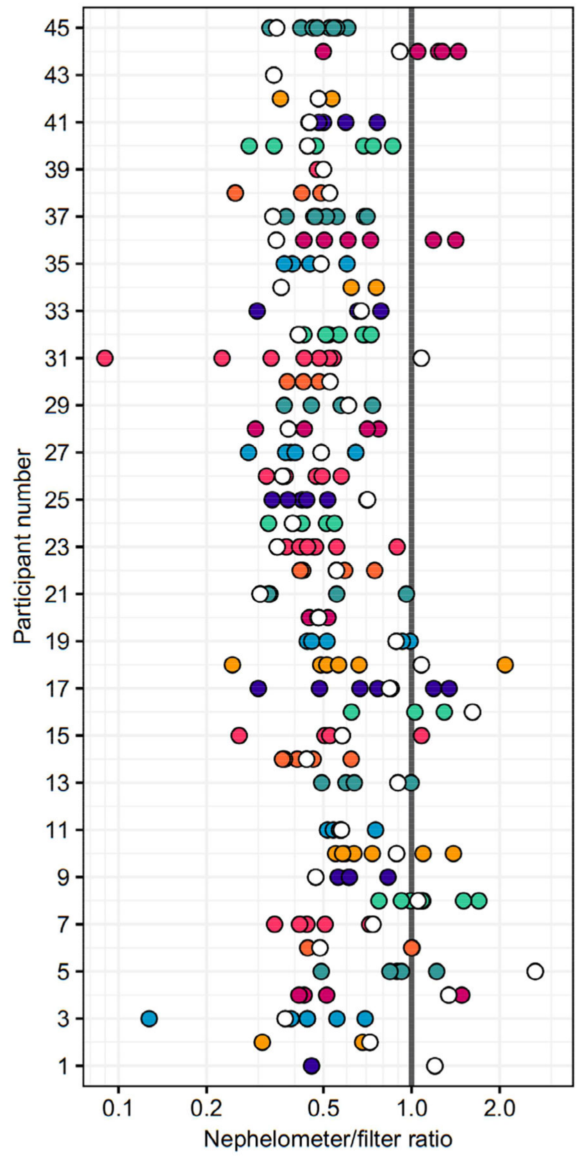


Figure 5.

Ratio of the 33-hour average nephelometer- and filter-derived $PM_{2.5}$ concentrations (n = 249) vs. participant number (n = 44). Marker colors are varied between participants to improve readability. The first sample collected by each participant is shown with a white marker. The solid vertical line represents a ratio of 1.

Table 1.

Summary of results from the main analysis and the sensitivity analyses (1st filtering step, 2nd filtering step, 3rd filtering step, 4th filtering step). For the main analysis, 10-second average nephelometer readings below the limit of detection (LOD) were adjusted to LOD/$\sqrt{2}$ before performing the relative humidity (RH) correction. For the sensitivity analyses, the RH correction was applied directly to the 10-second average nephelometer readings, without adjusting readings below the LOD. The first set of filtering criteria used in the sensitivity analyses were the same criteria used in the main analysis. The criteria used in the second, third, and fourth steps of the sensitivity analyses were progressively more stringent.

	Main analysis	**Sensitivity analyses: 1st filtering step**	**Sensitivity analyses: 2nd filtering step**	**Sensitivity analyses: 3rd filtering step**	**Sensitivity analyses: 4th filtering step**
Filtering criteria	Filter mass > LOD Filter conc. < 145 μg-m^{-3} pDR data available ≥ 85% pDR (10 s) < LOD → LOD/$\sqrt{2}$	Filter mass > LOD Filter conc. < 145 μg-m^{-3} pDR data available ≥ 85%	Filter mass > LOD Filter conc. < 145 μg-m^{-3} pDR data available ≥ 85% ≤ 25% pDR readings = 0 μg-m^{-3}	Filter mass > LOD Filter conc. < 145 μg-m^{-3} pDR data available ≥ 85% ≤ 25% pDR readings = 0 μg-m^{-3} ≤ 50% pDR readings below LOD	Filter mass > LOD Filter conc. < 145 μg-m^{-3} pDR data available > 85% ≤ 25% pDR readings = 0 μg-m^{-3} ≤ 50% pDR readings below LOD pDR 33-hour conc. > 4 μg-m^{-3}
No. samples	249	249	159	81	68
No. participants	44	44	42	35	31
Sample dates	100	100	86	57	51
$PM_{2.5}$ conc. (μg-m^{-3}) measured using filter					
Median	8	8	9	11	12
Interquartile range	6 – 11	6 – 11	6 – 13	8 – 19	9 – 24
Range	5 – 83	5 – 83	5 – 83	5 – 83	5 – 83
Correlation between nephelometer- and filter-derived conc.					
Spearman's rho	0.77	0.76	0.77	0.74	0.66
Difference between nephelometer- and filter-derived conc.					
Fraction ≤ |5| μg-m^{-3}	183/249 (73%)	157/249 (63%)	108/159 (68%)	50/81 (62%)	39/68 (57%)
Fraction ≤ 20%	32/249 (13%)	28/249 (11%)	25/159 (16%)	20/81 (25%)	20/68 (29%)
Median absolute difference (μg·m^{-3})	3.6	4.5	4.0	4.0	4.3
Median percent difference	49%	57%	50%	42%	36%
Nephelometer/filter ratio					
Median	0.52	0.44	0.52	0.68	0.74
25^{th} - 75^{th} percentile	0.43 – 0.70	0.29 – 0.68	0.38 – 0.77	0.49 – 1.0	0.54 – 1.0
10^{th} - 90^{th} percentile	0.35 – 1.1_	0.17 – 1.0_	0.29 – 1.1_	0.39 – 1.2	0.44 – 1.3

	Main analysis	Sensitivity analyses: 1st filtering step	Sensitivity analyses: 2nd filtering step	Sensitivity analyses: 3rd filtering step	Sensitivity analyses: 4th filtering step
Fraction = 0.8 – 1.2	32/249 (13%)	28/249 (11%)	25/159 (16%)	20/81 (25%)	20/68 (29%)
Fraction < 1	220/249 (88%)	222/249 (89%)	136/159 (86%)	61/81 (75%)	48/68 (71%)
Intraclass Correlation Coefficient					
Between participants (95% CI)	0.27 (0.15–0.42)	0.30 (0.18–0.45)	0.26 (0.10–0.44)	0.23 (0.00–0.49)	0.30 (0.18 – 0.45)
Between dates (95% CI)	0.14 (0.00–0.29)	0.07 (0.00–0.22)	0.20 (0.00–0.41)	0.00 (0.00–0.27)	0.07 (0.00 – 0.22)

Table 2.

Accuracy of uncorrected nephelometer-derived 33-hour personal $PM_{2.5}$ concentrations, relative to the filter-derived concentrations, compared to accuracy after correction using: (1) the median nephelometer/filter ratio calculated from a random subset of 44 samples, (2) the nephelometer/filter ratio calculated from the first sample for each of the 44 participants, and (3) factors predicted using linear models fit and tested using five-fold cross-validation. Model-predicted correction factors were only available for 245/249 samples because not all predictor variables were successfully measured during all samples.

Correction factor (CF) type	None	Median	Participant-specific	Model-predicted
Number of samples				
Used to calculate CF(s) or train model	-	44	44	≈200 per fold
Used to test CF(s) or Model	-	249	249	≈50 per fold
Total	249	249	249	245
Difference between nephelometer- and filter- derived concentrations				
Fraction ≤ |5 $\mu g \cdot m^{-3}$|	183/249 (73%)	188/249 (76%)	189/249 (76%)	189/245 (77%)
Fraction ≤ 20%	32/249 (13%)	106/249 (43%)	109/249 (44%)	103/245 (42%)
Median absolute difference	3.6 $\mu g \cdot m^{-3}$	2.0 $\mu g \cdot m^{-3}$	2.0 $\mu g \cdot m^{-3}$	2.0 $\mu g \cdot m^{-3}$
Median percent difference	49%	24%	25%	24%
Nephelometer/filter ratio				
Median	0.52	0.95	1.0	0.98
25^{th}–75^{th} percentile	0.43 – 0.70	0.78 – 1.3	0.77 – 1.3	0.77 – 1.3
10^{th}–90^{th} percentile	0.35 – 1.1	0.63 – 1.9	0.48 – 1.7	0.63 – 1.7