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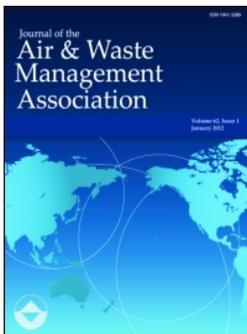
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TECHNICAL PAPER

Comparison of real-time instruments and gravimetric method when measuring particulate matter in a residential building

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ABSTRACT

This study used several real-time and filter-based aerosol instruments to measure PM_{2.5} levels in a high-rise residential green building in the Northeastern US and compared performance of those instruments. PM_{2.5} 24-hr average concentrations were determined using a Personal Modular Impactor (PMI) with 2.5 µm cut (SKC Inc., Eighty Four, PA) and a direct reading pDR-1500 (Thermo Scientific, Franklin, MA) as well as its filter. 1-hr average PM_{2.5} concentrations were measured in the same apartments with an Aerotrak Optical Particle Counter (OPC) (model 8220, TSI, Inc., Shoreview, MN) and a DustTrak DRX mass monitor (model 8534, TSI, Inc., Shoreview, MN). OPC and DRX measurements were compared with concurrent 1-hr mass concentration from the pDR-1500. The pDR-1500 direct reading showed approximately 40% higher particle mass concentration compared to its own filter (n = 41), and 25% higher PM_{2.5} mass concentration compared to the PMI_{2.5} filter. The pDR-1500 direct reading and PMI_{2.5} in non-smoking homes (self-reported) were not significantly different (n = 10, R² = 0.937), while the difference between measurements for smoking homes was 44% (n = 31, R² = 0.773). Both OPC and DRX data had substantial and significant systematic and proportional biases compared with pDR-1500 readings. However, these methods were highly correlated: R² = 0.936 for OPC versus pDR-1500 reading and R² = 0.863 for DRX versus pDR-1500 reading. The data suggest that accuracy of aerosol mass concentrations from direct-reading instruments in indoor environments depends on the instrument, and that correction factors can be used to reduce biases of these real-time monitors in residential green buildings with similar aerosol properties.

Implications: This study used several real-time and filter-based aerosol instruments to measure PM_{2.5} levels in a high-rise residential green building in the northeastern United States and compared performance of those instruments. The data show that while the use of real-time monitors is convenient for measurement of airborne PM at short time scales, the accuracy of those monitors depends on a particular instrument. Bias correction factors identified in this paper could provide guidance for other studies using direct-reading instruments to measure PM concentrations.

PAPER HISTORY

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Introduction

Over the past 15 years, studies have shown strong correlations between elevated particulate matter (PM) levels and a range of health effects, including early mortality (Schwartz et al., 1996; Klemm and Mason, 2000; U.S. EPA, 1996), exacerbation of respiratory-tract disease, reduced lung function (Hoek et al., 1998), and cardiovascular disease (Peters et al., 2000; Pope et al., 1999). According to the EPA, every 10-µg/m³ increase in fine particles (<2.5 µm in diameter) produces a 6% increase in the risk of death from cardiopulmonary disease and increased morbidity among all age groups

(U.S. EPA, 1996). Among exposed populations, children are more susceptible than adults to many health effects caused by air pollution (Mathieu-Nolf, 2002). It was reported that every 10-µg/m³ increase in PM₁₀ particles (<10 µm in aerodynamic diameter) is associated with a 10% decrease in children's peak expiratory flow (Dockery and Pope, 1994). Such effects are especially severe when both indoor and outdoor concentrations of pollutants including PM₁₀, NO₂, SO_x, and CO are elevated (Mathieu-Nolf, 2002).

Health effects of airborne particulate matter and other pollutants present indoors are important because

people spend about 90% of their time inside buildings, and most of that time is in residences (Klepeis et al., 2001; Spengler et al., 1985; Spengler and Sexton, 1983). Green building practices are increasingly being used in residential buildings to reduce environmental burdens associated with building construction and operation (Eichholtz, Kok, and Quigley, 2013). Both the U.S. EPA and real-estate corporations have recognized the importance of indoor air quality in green buildings (Albanese Organization, 2004; U.S. EPA, 2009). A growing body of research suggests that green buildings have better indoor environmental (e.g., air quality, thermal comfort) quality and occupant satisfaction, productivity, and health than nongreen buildings (Abbaszadeh et al., 2006; Ahrentzen et al., 2012; Eichholtz, Kok, and Quigley, 2013). At the same time, data on PM in residential green buildings are only beginning to appear in the peer-reviewed literature and control of indoor PM concentrations is proposed in few green building certifications (Colton et al., 2014; Patton et al., 2016; Wei, Ramalho, and Mandin, 2015). The construction and operation of individual green buildings, as well as occupant behaviors, may strongly affect measured mass concentrations and indoor/outdoor ratios of PM (Patton et al., 2016).

Both filter-based gravimetric and real-time methods have been used to measure PM in buildings (e.g., Ramachandran et al., 2000; Wallace et al., 2006; Wallace et al., 2011; Wheeler et al., 2011; Yanosky, Williams, and MacIntosh, 2002; Van Ryswyk et al., 2014; Osman et al., 2007). Filter-based methods are robust and relatively easy to operate, but the accumulation of sufficient particle mass and filter weighing usually take a considerable amount of time (McMurry, 2000; Pui, 1996). In addition, conditioning of filters to remove moisture can also cause the loss of potentially health-related volatile compounds (Huang et al., 2013). Many real-time monitors for particulate mass concentrations sense light reflected by a particle stream onto a photodetector and convert the photometric voltage output into concentrations via a calibration curve (Wang et al., 2009). Photometers (e.g., DustTrak) measure light reflected at a specific angle, while nephelometers (e.g., pDR) measure light reflected over as wide a range of angles as possible (Hinds, 1999). Another monitor similar to a photometer, the optical particle counter, illuminates a single particle at a time to count the number of particles in defined size bins (Hinds, 1999). Real-time particle monitors can measure PM concentration spikes and peak exposures, which could have importance for health effects (McMurry, 2000; Jiang et al., 2011). However, the relationships of real-time measurements to the gravimetric

measurements that are used to set air quality standards are often unknown because the relationship between light scattering intensity (and therefore instrument response of optical particle counters or photometers) and particle mass concentration determined gravimetrically depends on properties of the aerosols (e.g., density, reflectivity, size, shape, and composition) used for calibration and the aerosols in the environment (e.g., Kim et al., 2004; Yanosky, Williams, and MacIntosh, 2002; O'Shaughnessy and Slagley, 2002; Pui, 1996; Wang et al., 2009; Wallace et al., 2011; Sorensen et al., 2011; Liu and Daum, 2000; McMurry, 2000; Chang et al., 2001).

In the field, real-time monitors are typically calibrated by co-locating real-time and gravimetric instruments, and by using a scaling factor (i.e., calibration factor) or curve of best fit to adjust the measurements reported by the real-time monitors to match the gravimetric measurements (e.g., Sioutas et al., 2000; Wallace et al., 2011; Wang et al., 2009). For example, residential measurements of PM_{2.5} using a personal DataRAM sampling passively (pDR-1000, Thermo Scientific, Inc.) were 1.10–1.92 times higher than concurrent measurements with Harvard Impactors (HI) and Personal Environmental Monitors (PEM), two types of portable gravimetric instruments (Howard-Reed et al., 2000; Wallace et al., 2006; Wallace et al., 2011). Actively sampling DustTraks (a photometer; models 8520 and earlier) measured concentrations that were 1.94–2.57 times higher than filter-based and federal reference method measurements in occupied and test homes (Ramachandran et al., 2000; Wallace et al., 2011; Yanosky, Williams, and MacIntosh, 2002). All of these comparisons resulted in high correlations (i.e., >0.77) among measurements using different methods. Wheeler et al. (2011) demonstrated that correcting different monitors for site-specific variation in instrument response could lead to high correlation (slope = 1.02; $R^2 = 0.712$) and insignificant bias (intercept = $-0.58 \mu\text{g}/\text{m}^3$) between different real-time monitors and filtration-based measurements (Wheeler et al., 2011), thus allowing for faster and more convenient estimation of indoor PM. However, the exact relationship between different methods depends on the particles and environments being sampled. Wallace et al. (2006) found that the pDR-1000 had larger overestimates relative to HI and PEM filters for particles from cooking than for particles related to cleaning and personal care products. A study of another photometer (i.e., TSI SidePak) demonstrated that real-time monitors overestimated gravimetric measurements of PM_{2.5} by factors of 0.92–1.8 for outdoor sources, 1.3 for toasting bread, and 3.4 for cigarette smoke (Jiang

et al., 2011). Therefore, factors specific to residential indoor air are needed for real-time monitors to approximate actual PM mass concentrations.

In our recent study investigating a green building, a multidisciplinary research team collaborated on a post-occupancy evaluation of the building to examine the advantages, synergies (positive and negative), and trade-offs for the environmental and social impact of a green multifamily dwelling on its residents. As part of the study, we used different impactors equipped with filters and real-time particle monitors to determine relationships in PM_{2.5} concentrations measured by those instruments. The objective of this paper is to describe the relationships among measurements by the various portable monitors.

Materials and methods

Study site and time frame

The study was performed in both wings of a two-wing multi-apartment (127) residential building in the northeastern United States. The six- and seven-story complex was the largest multifamily high rise building in the nation with Federal Energy Star certification at the time of the study. The building was naturally ventilated except for exhaust vents (designed for 50 m³/hr) in the kitchen and bathroom of each apartment. Smoking was allowed in apartments but not in the common spaces of the buildings, and occupants were allowed to cook, clean, and open windows. The building is located about 100 m from an elevated rail line and at the nexus of two side streets in an urban community known for a predominance of truck routes.

All building occupants were invited to participate in the study via communications by management and the research team using flyers and meetings. Forty households (mostly two-bedroom and one three-bedroom) volunteered for the study and were administered a baseline questionnaire examining their health, lifestyle, and satisfaction with the building. Twenty-one households (apartments) with presence of asthma, obesity, smoking, or a combination of these as per questionnaire were selected for and agreed to additionally participate in detailed indoor air quality (IAQ) measurements. On each sampling day, IAQ measurements were made in one to three apartments (usually in the sitting room). The IAQ measurements were performed in three phases, corresponding to the phases of a cleaning behavioral intervention: Phase I: July–September 2011, Phase II: October–December 2011, and Phase III: March–May 2012. In Phase I, 21 apartments were sampled, while in Phases II and III, 17 apartments were

sampled due to participant attrition. These 17 apartments participated in Phase I sampling as well.

Equipment used

Twenty-four-hour PM_{2.5} samples were collected by a Personal Modular Impactor (PMI) and a personal DataRAM (pDR-1500) set up side-by-side on a tripod. The PMI (PMI_{2.5}; SKC, Inc., Eighty Four, PA) is a single-stage impactor equipped with a filter that uses a 25-mm pre-oiled impaction disc to remove particles larger than 2.5 µm and reduce particle bounce. An AirChek XR5000 Pump (SKC, Inc.) provided the sampling airflow of 3 L/min through the PMI_{2.5}. The SKC PM_{2.5} impactor, part of SKC's modular impactor line, closely follows the EPA's PM_{2.5} curve (Trakumas, Smith, and Nachreiner, 2010). Collocated runs of impactors from this series with MiniVol (Airmetrics, Inc.) and FH 62 C14 continuous ambient PM monitor (Thermo Andersen) samplers showed very good agreement (Trakumas and Smith, 2008; Trakumas et al., 2006).

The pDR-1500 (Thermo Scientific, Franklin, MA) is a sensitive nephelometric monitor with a cyclone inlet for measurement of the respirable fraction of the airborne particulate matter in different environments. The pDR-1500 reported the average PM_{2.5} concentration every 10 min and also collected particles that passed through the sensing zone on a 37-mm filter for gravimetric analysis. The pDR-1500 was operated at a flow rate of 1.52 L/min for a PM_{2.5} cut size. 37-mm Teflon filters with 2.0 µm pore size (SKC, Inc.) were used for both the PMI_{2.5} sampler and the pDR-1500. Sampling time was programmed into the sampling pump of the PMI_{2.5} and the pDR-1500 control software. Sampling flow rates were verified at the start and end of sampling using a mass flowmeter (model 4140, TSI, Inc., Shoreview, MN), and the average flow rate was used to calculate the amount of air sampled. All filters were allowed to equilibrate in a weighing room with controlled temperature (21°C) and relative humidity (30–40%) for a minimum of 72 hr prior to weighing before and after sampling. Filters were weighed using a Mettler Toledo MT5 Microbalance. All weighing sessions included weighing of 50- and 200-µg standards. In addition, three blank filters were always kept in the weighing room and weighed during each session as part of the quality assurance (QA) procedure. Their weight changes were within 2–3 µg and no drift was observed. All field sample masses were substantially above the detection limit.

In addition to 24-hr average $PM_{2.5}$ concentration, various particle mass and size fractions were measured for 1 hr using two instruments: DustTrak DRX (TSI 8534, TSI, Inc.) and AeroTrak optical particle counter (OPC, model 8220, TSI, Inc.). The DustTrak DRX is a combined photometer and optical counter, where a laser diode illuminates a sample stream and reflected light is directed onto a photodetector by a mirror. When properly calibrated the DustTrak DRX has <10% error in comparison to TEOM measurements (Wang et al., 2009). The photometric voltage output from the DustTrak DRX was converted to real-time particulate matter (PM) mass concentrations corresponding to PM_1 , $PM_{2.5}$, PM_4 , PM_{10} , and total PM size fractions. The OPC provided particle number concentrations in the following diameter ranges: 0.3–0.5, 0.5–1.0, 1.0–2.5, 2.5–5.0, 5.0–10.0, and >10.0 μm . To compare the 1-hr average mass concentrations with other instruments, the number concentrations measured by OPC were converted into mass concentration using the following assumptions: (1) Particles were spherical, (2) particles in a particular size channel had a size equivalent to the arithmetic mean of that channel, and (3) indoor particles had density of 1.65 g/cm^3 (Tittarelli et al., 2008). Except for at concentrations <0.5 $\mu\text{g}/\text{m}^3$ (poor DustTrak signal) or > 100 $\mu\text{g}/\text{m}^3$ (OPC coincidence error), the DustTrak DRX and OPC have good agreement in measurements of monodisperse aerosols (Wang et al., 2009). These two instruments were operated side-by-side on a table in the same sitting room. During this time, the $PMI_{2.5}$ and pDR-1500 were also running.

All real-time monitors were manufacturer-calibrated for the study. As per manufacturers, the AeroTrak 8220 was calibrated using NIST traceable polystyrene latex (PSL) spheres; the DustTrak DRX was calibrated using emery oil and nominally adjusted to the respirable mass of standard ISO 12103-1, A1 test dust (Arizona Road Dust); and the pDR-1500 was calibrated using Arizona Road Dust. PSL has density $\rho_p = 1.05 \text{ g}/\text{cm}^3$ and refractive index $m = 1.59$, while Arizona Road Dust has density $\rho_p = 2.65 \text{ g}/\text{cm}^3$ and refractive index $m = 1.54$ (refractive index of Arizona Road Dust; Applied Physics, Inc., 2015; Wang et al., 2009).

Laboratory collocation tests were done where possible to assess the precision of the monitors. Two co-located PMI impactors were operated over 24 hr on 7 different days in a low-pollution environment ($\sim 0.06 \text{ mg}$ collected on average per 24 hr); their results were not statistically different ($p = 0.09$ from a two-tailed t -test). The mass collected by two co-located SKC modular impactors in a high concentration environment ($\sim 0.8 \text{ mg}$ on average in 24-hr samples) differed by less than 5% and were not statistically different ($p = 0.49$ from a two-tailed t -test).

Two TSI OPCs were run side by side as part of this project, and particle counts within individual channels showed good agreement (average R^2 of six size channels = 0.85). The two pDRs used in this study were run side-by-side indoors at 1.2 L/min and 3 L/min sampling flow rates, and the average ratios of their measured values were 1.02 ± 0.13 and 1.06 ± 0.09 , respectively. The Pearson correlations for those two flow rates were 0.73 and 0.88, respectively. No collocation test was done for the DustTrak DRX because only one monitor was available; the manufacturer reported sampled airflow accuracy was $\pm 5\%$ (TSI, Incorporated, 2014).

Statistical analysis

STATISTICA 7 software package (StatSoft, Inc., Tulsa, OK) was used to evaluate both constant and proportional bias and to assess the correlation between any two sampling methods. A significance level of 0.05 was used in all tests. Ordinary least-squares linear regression was performed for the $PM_{2.5}$ levels obtained by two sampling methods. The regression is presented by the following equation:

$$y = \beta_1(\pm SE)x + \beta_0(\pm SE) \quad (1)$$

where β_1 and β_0 are regression coefficients, and SE is the standard error of β values. If two sampling methods match exactly, the regression intercept, β_0 , should be equal to 0 and the regression slope, β_1 , should be equal to 1. Thus, the following hypotheses were developed and p -values for each hypothesis were calculated to compare sampling methods:

$$\text{Null hypothesis 1 : } \beta_1 = 1, \text{ reject if } p_1 < 0.05 \quad (2)$$

$$\text{Null hypothesis 2 : } \beta_0 = 0, \text{ reject if } p_2 < 0.05 \quad (3)$$

If the regression intercept, β_0 , was found to be significantly different from zero, it was considered to indicate the systematic (constant) bias of the $PM_{2.5}$ levels between two samplers or methods. If the regression slope, β_1 , was found to be significantly different from unity (1), it was considered to indicate the proportional bias of $PM_{2.5}$ levels between two samplers. The coefficient of determination (R^2) was used to describe the correlation of measured levels between two sampling methods, and the root mean square error (RMSE) was calculated to compare the relative agreement of the sampling methods.

Results and discussion

The data from all three sampling phases were pooled together for the comparison analysis, for a total of 48 sets of measurements in 21 apartments. The 24-hr $PMI_{2.5}$, OPC, and DustTrak DRX records were

complete; only 40 apartment visits had pDR-1500 data because only two pDR-1500 units were available (Table 1). For 1-hr average PM_{2.5} levels, the OPC had only 34 valid data points from Phases II and III due to the malfunction (faulty data retrieval) of the unit in Phase I. Most measurement methods recorded mean PM_{2.5} mass concentrations between 22 µg/m³ and 35 µg/m³, although the mean OPC measurement was much lower (5.96 µg/m³). The highest recorded PM_{2.5} mass concentration was 156 µg/m³, and was obtained from a pDR-1500 direct reading. Indoor and outdoor measurements from the DustTrak DRX on the same day showed that concentrations of PM_{2.5} were typically higher indoors than outdoors; ratios of indoor concentrations to outdoor concentrations (both measured by DustTrak DRX) ranged from 0.53 to 9.26 (mean = 2.03, median = 1.41) (Patton et al., 2016). During measurements, investigators observed open windows in 25 apartments, closed windows in 16 apartments, and window air-conditioning units in 10 apartments. Investigators reported cooking in five nonsmoking apartments and smoke infiltration from a neighboring apartment in one nonsmoking apartment. One apartment with a smoker also had oil fragrance burning. No other sources of combustion were observed in the apartments.

A comparison of the 24-hr PM_{2.5} average concentrations between pDR-1500 reading and pDR-1500 filter measurements is presented in Figure 1. The intercept was not significantly different from zero ($\beta_0 = -3.30, p = 0.233$), but the slope was significantly different from unity ($\beta_1 = 1.42, p < 0.001$). Therefore, there was a significant proportional bias between these two methods using the same instrument, although the concentration values provided by the two methods were correlated ($R^2 = 0.804$). The value $\beta_1 = 1.42$ also indicates that direct reading showed concentrations on average 42% higher than filter weighing. One possible explanation for this result is that some PM was lost from the pDR filter during sampling, transport, or filter weighing (Alderman and Ingebretsen, 2011; Scian et al., 2009). Alternatively, as mentioned on the calibration certificate, it is possible the pDR-1500 calibration factor determined by calibration with Arizona Road Dust was not the best fit for the environment investigated in this study. The pDR-1500 can save

up to 50 different calibration factors, so saving a calibration factor in the instrument software based on preliminary data comparing the pDR-1500 direct reading to either the pDR-1500 filter or another gravimetric measurement can improve the accuracy of the direct readings from the pDR-1500 (Thermo Fisher Scientific, Inc., 2014).

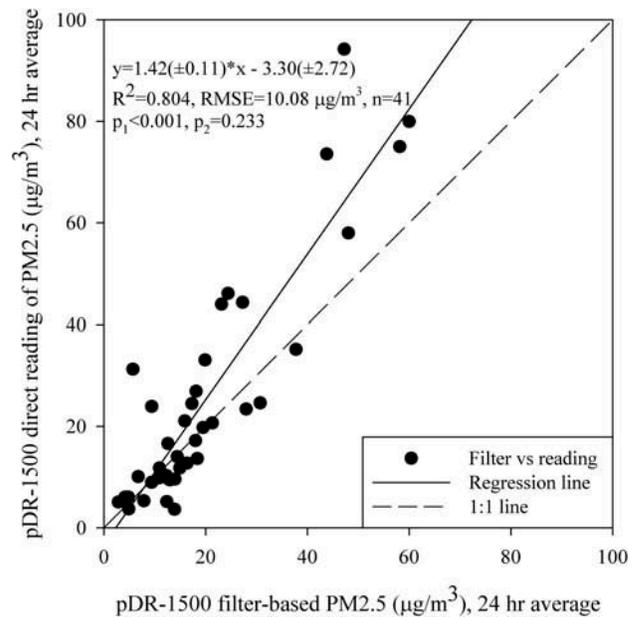


Figure 1. Comparison of PM_{2.5} 24 hr average concentrations between pDR-1500 direct reading and pDR-1500 filter-based measurements. The regression is presented by the following equation: $y = \beta_1(\pm SE)x + \beta_0(\pm SE)$ where β_1 and β_0 are regression coefficients, and SE is the standard error of β values.

The PMI_{2.5} (filter) comparison with pDR-1500 filter measurements is shown in Figure 2. The intercept was not significantly different from zero ($\beta_0 = 0.04, p = 0.982$) but the slope was significantly smaller than unity ($\beta_1 = 0.79, p = 0.002$); that is, on average pDR-1500 filter underestimated PM_{2.5} concentration by approximately 20% compared to the PMI_{2.5} filter. The comparison of the 24-hr average PM_{2.5} levels between PMI_{2.5} filter and pDR-1500 direct reading measurements is shown in Figure 3. The intercept was significantly different from zero ($\beta_0 = -6.38, p = 0.042$) and the slope was significantly different from one ($\beta_1 = 1.25, p = 0.018$). Therefore, there were significant

Table 1. Summary of PM_{2.5} measurements (µg/m³) from the different real-time and filter-based metrics.

Measurement type	Monitor	Temporal resolution	n	Mean	Median	Minimum	Maximum
Direct reading	DustTrak DRX	1 min*	48	32.74	25.66	8.30	138.02
	OPC	1 min*	34	5.96	4.01	0.59	41.66
	pDR-1500 direct reading	10 min	40	27.61	20.25	3.70	156.00
Filter-based	pDR-1500 filter	24 hr	40	22.30	17.64	2.80	76.80
	PMI _{2.5} impactor	24 hr	48	25.92	18.49	2.80	113.08

*Each 1-min measurement from these monitors consists of 60 sec of measurement and a 10-sec break.

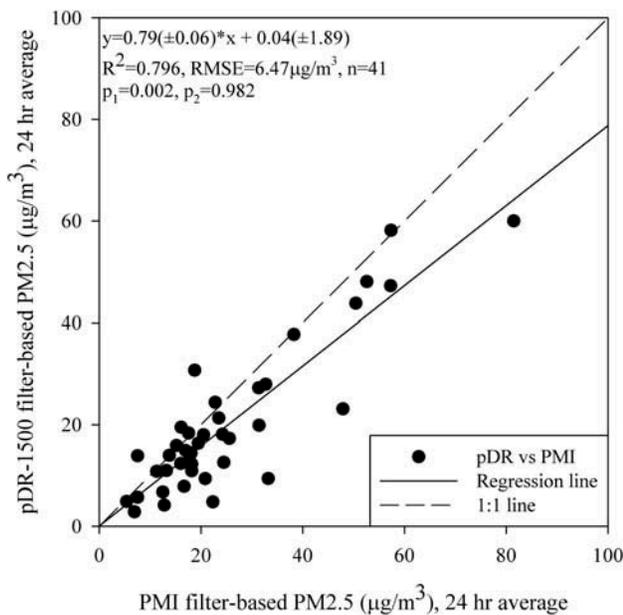


Figure 2. Comparison of $PM_{2.5}$ 24-hr average concentrations determined by PMI $PM_{2.5}$ filter-based measurements and pDR-1500 filter-based measurements. The regression is presented by the following equation: $y = \beta_1(\pm SE)x + \beta_0(\pm SE)$ where β_1 and β_0 are regression coefficients, and SE is the standard error of β values.

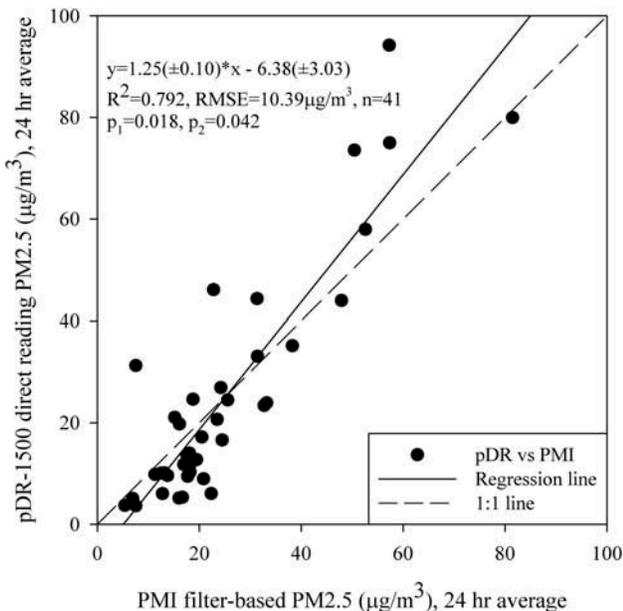


Figure 3. Comparison of $PM_{2.5}$ 24-hr average concentrations determined by PMI $PM_{2.5}$ filter-based measurements and pDR-1500 direct reading measurements. The regression is presented by the following equation: $y = \beta_1(\pm SE)x + \beta_0(\pm SE)$ where β_1 and β_0 are regression coefficients, and SE is the standard error of β values.

systematic ($6.38 \mu\text{g}/\text{m}^3$) and proportional (25%) biases between these two methods, with pDR-1500 showing higher concentrations compared to $PMI_{2.5}$ impactor.

Since real-time monitors are known to overestimate concentrations of smoke particles more than other indoor aerosols relative to gravimetric measurements (Jiang et al., 2011), we conducted stratified analyses by household smoking status. According to the questionnaire data administered to the study participants, about 62% of apartments participating in this study self-reported having one or more smokers in the apartment. No smoking was observed during the time that investigators were in the apartments (except for one case of smoking in a neighboring apartment), and the questionnaire did not ask for the frequency or room of smoking. Thus, the data of the 24-hr $PM_{2.5}$ average concentrations measured by $PMI_{2.5}$ filter and pDR-1500 direct reading were stratified into two groups according to the self-reported smoking and nonsmoking status in investigated homes, and the results are presented in Figure 4. It can be seen that for nonsmoking homes the correlation between these two methods has improved ($R^2 = 0.937$) and that the biases were reduced to not statistically significant levels ($\beta_0 = -1.78$, $p = 0.604$; $\beta_1 = 1.04$, $p = 0.718$). On the other hand, for smoking homes, the differences between these two methods increased ($R^2 = 0.773$; $\beta_0 = -10.29$, $p = 0.015$; $\beta_1 = 1.44$, $p = 0.005$). While direct comparison of the PMI or pDR-1500 filter concentrations to the OPC measurements was not possible because the monitors were not operating for the same amount of time, we would expect similarly improved relationships between these measurement techniques for subsets of homes with similar aerosol composition.

The differences in monitor comparisons between smoking and nonsmoking apartments may have been related to different factors affecting light-based and filter-based measurements, and are similar to differences previously reported in chamber studies of environmental tobacco smoke that compared responses of nephelometers to gravimetric measurements (Brauer et al., 2000; Jenkins et al., 2004). In general, filter-based readings are probably more accurate than light-based readings because the output of direct reading instruments depends on the reflective properties and size distribution of particles that were used for calibration (Liu and Daum, 2000; Sioutas et al., 2000; Kim et al., 2004; Yanosky, Williams, and MacIntosh, 2002; O'Shaughnessy and Slagley, 2002; Pui, 1996; Wang et al., 2009; Jiang et al., 2011). Although the mass median diameter of environmental tobacco smoke (ETS) is $\sim 200 \text{ nm}$ (Klepeis et al., 2003), larger than the lower limit of 100 nm for photometers that rely on Mie scattering, there may have been some undercounting of the smallest particles. Photometers calibrated with Arizona Road Dust are expected to

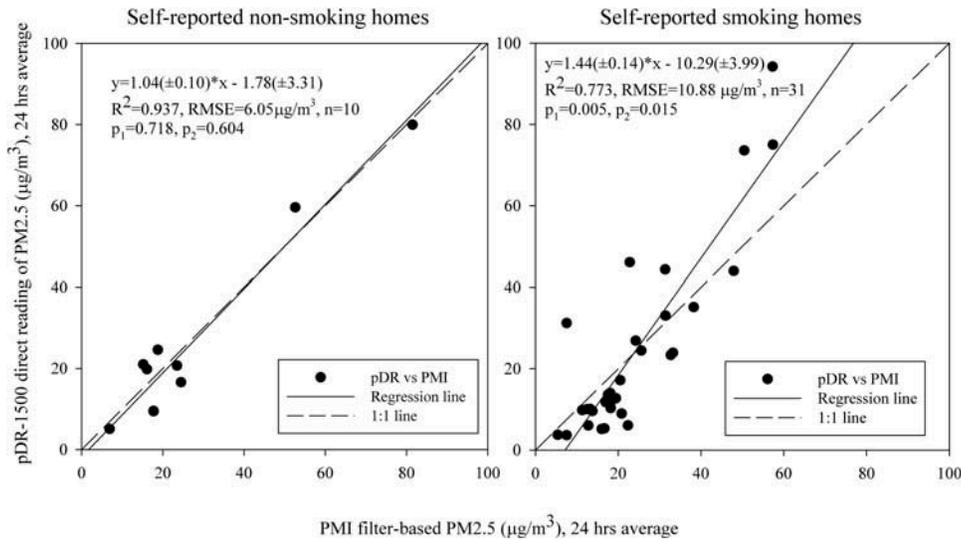


Figure 4. Comparison of PM_{2.5} 24 hr average concentrations determined by PMI PM_{2.5} filter-based measurements and pDR-1500 direct reading measurements, with data stratified according to the self-reported smoking status in investigated apartments. The regression is presented by the following equation: $y = \beta_1(\pm SE)x + \beta_0(\pm SE)$ where β_1 and β_0 are regression coefficients, and SE is the standard error of β values.

read high for ETS because smoke particles are both smaller and less dense than Arizona Road Dust (Jiang et al., 2011). However, it has also been observed that filter-based techniques can have inaccuracies in both mass and composition due to the loss of volatile and semivolatile components of tobacco smoke when sampling smoking emissions (Thorne and Adamson, 2013; Adamson et al., 2012; Alderman and Ingebrethsen, 2011; Scian et al., 2009). Thus, it is possible that some particles originating from tobacco smoke lost mass due to evaporation during sampling, as well as during filter handling and weighing, and that this loss caused differences between gravimetric (PMI_{2.5} and pDR-1500 filters) and real-time measurements. It is also possible that the composition of particles in smoking and nonsmoking households had different light-scattering properties and thus affected pDR-1500 readings. Thus, according to our data, the discrepancy between pDR-1500 direct reading and the PMI_{2.5} filter method could be at least partially explained by the presence/absence of tobacco smoke and different properties of particles in those two household types. At the same time, due to the relatively low number of data points, especially for non-smoking homes, the conclusion should be treated with caution and a more comprehensive investigation is warranted.

Another set of comparisons was carried out for 1-hr concentrations measured by direct reading instruments. The PM_{2.5} mass estimate by AeroTrak OPC was compared with a corresponding 1-hr average of pDR-1500

readings and the data are presented in Figure 5. The correlation between these two methods was high with $R^2 = 0.936$. However, the slope ($\beta_1 = 0.40$) of the regression line was significantly lower than unity ($p_1 < 0.001$), so the OPC PM_{2.5} mass estimate

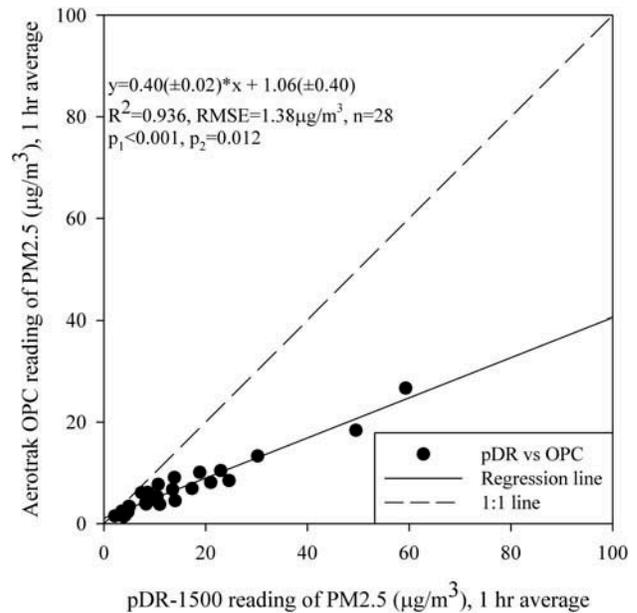


Figure 5. Comparison of PM_{2.5} 1-hr average concentrations determined by pDR-1500 direct reading measurements and AeroTrak OPC estimate assuming particles are spherical and have a density of 1.68 g/cm³. The regression is presented by the following equation: $y = \beta_1(\pm SE)x + \beta_0(\pm SE)$ where β_1 and β_0 are regression coefficients, and SE is the standard error of β values.

underestimated that based on pDR-1500 by a factor of 2.5. This underestimation is likely to be caused by the coincidence losses inside the OPC and/or violation of one or more of the assumptions used to calculate particles mass concentration based on their number distribution. Because OPCs measure optical equivalence size relative to the polystyrene latex spheres (PSL) used for calibration, which have a higher refractive index (1.59) than most atmospheric aerosols, the measured optical size would likely underestimate geometric size, causing the mass to be further underestimated (Binnig, Meyer, and Kasper, 2007; Liu and Daum, 2000). Another possible contribution to a lower mass measured by the OPC could be nonsphericity of particles or the presence of high concentrations of particles smaller than the OPC's detection limit of 0.3 μm . Such particles were not measured in this study, but instances of their high concentrations indoors have been confirmed in an ongoing study (unpublished data). Systematic or constant bias was only $\sim 1 \mu\text{g}/\text{m}^3$, but it was statistically significant ($p = 0.012$). On the other hand, a strong correlation between these two methods suggests that the biases can be corrected by using the slope and intercept from a regression line of the OPC estimation versus the pDR-1500 direct reading (considered a reference here). This is a reasonable assumption given the absence of filter data for 1-hr of sampling and a relatively good agreement between pDR-1500 direct reading and its filter data. Such regression equation would be as follows:

$$y = 2.37x - 1.58 \quad (4)$$

where y equals the corrected value for the OPC mass estimation and x equals the uncorrected OPC mass estimation using the three assumptions already described.

A comparison of 1-hr average levels between DustTrak DRX $\text{PM}_{2.5}$ reading and pDR-1500 direct reading measurements is shown in Figure 6. The intercept of the regression line was significantly higher than zero ($\beta_0 = 10.46$, $p < 0.001$) and the slope was significantly higher than 1 ($\beta_1 = 1.94$, $p < 0.001$), indicating significant systematic and proportional biases between these two methods. The DRX appears to overestimate the mass concentration by approximately a factor of 2 and has a systematic bias of approximately $10 \mu\text{g}/\text{m}^3$ compared to pDR-1500 direct reading. The bias observed in the intercept could have been related to nonlinearities in the relationship between measures at very low concentrations, or related to how particles of different composition affected the reading. Both monitors read zero during zero-filter checks prior to measurements. According to Figure 3, the pDR-1500 direct

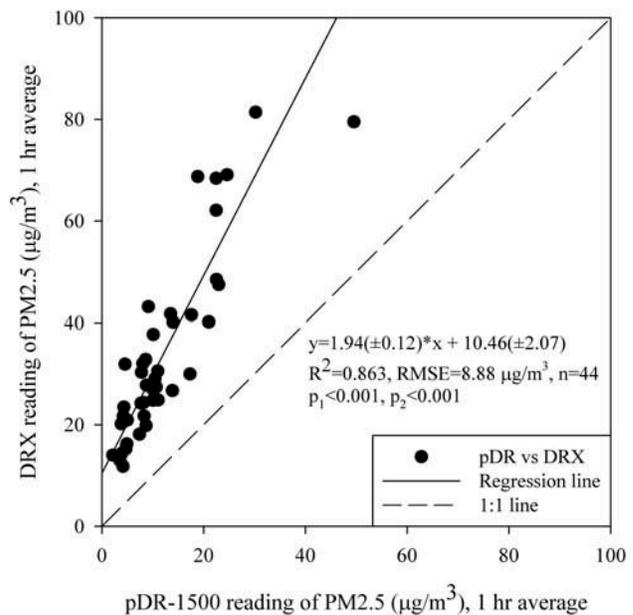


Figure 6. Comparison of $\text{PM}_{2.5}$ 1-hr average concentrations determined by pDR-1500 and DustTrak DRX direct reading measurements. The regression is presented by the following equation: $y = \beta_1(\pm\text{SE})x + \beta_0(\pm\text{SE})$ where β_1 and β_0 are regression coefficients, and SE is the standard error of β values.

reading overestimated mass concentration by 25% compared to PMI $\text{PM}_{2.5}$ values; thus, one can infer that the DustTrak DRX $\text{PM}_{2.5}$ reading overestimated $\text{PM}_{2.5}$ mass by a factor of 2.4 compared to the PMI filter. Although we used a different DustTrak model (DRX vs. 8520) in our study, the slope of the regression in the present study was near the middle of the range of those for previous indoor air studies comparing DustTraks to filter-based methods: 1.94 observed by Ramachandran et al. (2000) in the Minneapolis–St. Paul, MN, metropolitan area, 2.57 observed by Yanosky, Williams, and MacIntosh (2002) in Athens, GA, 2.78 observed by Wallace et al. (2011) in Windsor, ON, Canada, and 3 observed by Osman et al. (2007) in northeast Scotland.

The high correlation ($R^2 = 0.863$) between pDR-1500 and DustTrak DRX readings in our study indicates that their readings could be correlated by a linear regression function:

$$y = 0.44x - 2.86 \quad (5)$$

where y equals the corrected value for the DRX readings and x equals the uncorrected DRX readings. Therefore, our data support a calibration factor (inverse of overestimation factor) of 0.44 for the DustTrak DRX in the green building visited in this study, which is also similar to the manufacturer-recommended default of 0.38 (TSI, 2013). Using eq 5, one could quickly adjust

DustTrak DRX readings to more accurately reflect PM_{2.5} concentration indoors, if pDR-1500 direct readings were used as a reference.

We have introduced additional relationships among real-time particle monitors that can be applied as correction factors to estimate airborne PM_{2.5} concentration. Overall, our data show that most of the compared instrument pairs had systematic and proportional biases relative to each other; however, in most cases they agreed within a factor of 2. The largest discrepancy was observed between the pDR-1500 direct reading and the Aerotrak OPC. In general, our comparisons of real-time and filter-based measurements are in agreement with results reported in other studies (e.g., Howard-Reed et al., 2000; Wallace et al., 2006; Chang et al., 2001; Ramachandran et al., 2000; Wallace et al., 2011). Our comparisons may have been marginally affected by particle size and number concentration, and were probably not affected by humidity. Concentrations reported by the pDR-1500 and DustTrak DRX were weakly negatively correlated with the volume mean diameter reported by the OPC ($r = -0.3$ and $p = 0.06$ for both). Similarly, the nephelometer and photometer reported slightly lower concentrations when the OPC number mean was larger ($r = -0.3$, $p = 0.08$ for pDR-1500 and $r = -0.2$, $p = 0.18$ for DustTrak DRX). Although the humidity inside the building was higher than outdoors during the summer and lower than outdoors during the heating season, we did not observe associations between humidity and real-time pDR or DustTrak DRX measurements (Patton et al., 2016). While readings of optical monitors are known to be substantially affected by relative humidity levels >80% (Wallace et al., 2011, Fischer and Koshland, 2006), the range of relative humidity experienced during measurements (13–68%) would be unlikely to affect readings by more than ~5% (Yu et al., 2015), a difference much smaller than other differences among apartments in the study.

Conclusions

For airborne particulate matter monitoring, researchers have been using gravimetric methods in both indoor and outdoor environments. With the advance of real-time measurement instruments, it is now possible to measure PM dynamics in indoor and outdoor environments at short time scales and thereby obtain a detailed picture of PM_{2.5} variability, especially in health effects studies. At the same time, our data suggest that caution is needed when real-time aerosol monitors are used for determining mass concentration.

We found that measurements by pDR-1500 direct readings matched fairly well with gravimetric measurements from its own filter and PM_{2.5} sampler. The correlation between the two instruments especially improved when samples from nonsmoking households were considered (no significant bias and $R^2 = 0.937$). We also found that 1-hr Aerotrak OPC and DustTrak DRX measurements had significant and substantial systematic and proportional biases compared to pDR-1500 1-hr readings. OPC underestimated mass concentration by a factor of 2.5, while DustTrak DRX overestimated mass concentration by a factor of 2 compared to pDR-1500. However, these biases could be reduced for aerosols with similar properties, for example, in residential green buildings, by using correction factors determined by preliminary filter-based sampling in the environment to be monitored with real-time monitors. Additional studies of indoor PM_{2.5} particle shape and density are needed to develop more universally applicable conversions among different types of measurements. While it is unlikely that the exact same factors presented in this paper could be applied in other indoor studies given site-to-site variability of particle composition, we hope that our data will provide guidance for other studies using direct-reading instruments to measure concentrations of PM fractions.

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