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Effects of Aerosol Particle Size on the Measurement of Airborne PM_{2.5} with a Low-Cost Particulate Matter Sensor (LCPMS) in a Laboratory Chamber

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Abstract

Previous validation studies found a good linear correlation between the low-cost particulate matter sensors (LCPMS) and other research grade particulate matter (PM) monitors. This study aimed to determine if different particle size bins of PM would affect the linear relationship and agreement between the Dylos DC1700 (LCPMS) particle count measurements (converted to PM_{2.5} mass concentrations) and the Grimm 11R (research grade instrument) mass concentration measurements. Three size groups of PM_{2.5} (mass median aerodynamic diameters (MMAD): <1 μ m, 1 – 2 μ m, and >2 μ m) were generated inside a laboratory chamber, controlled for temperature and relative humidity, by dispersing sodium chloride crystals through a nebulizer. A linear regression comparing 1-min average PM_{2.5} particle counts from the Dylos DC1700 (Dylos) to the Grimm 11R (Grimm) mass concentrations was estimated by particle size group. The slope

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for the linear regression was found to increase as MMAD increased ($<1\mu\text{m}$: 0.75 ($R^2=0.95$), $1 - 2\mu\text{m}$: 0.90 ($R^2=0.93$) and $>2\mu\text{m}$: 1.03 ($R^2=0.94$). The linear slopes were used to convert Dylos counts to mass concentration and the agreement between converted Dylos mass and Grimm mass was estimated. The absolute relative error between converted Dylos mass and the Grimm mass was smaller in the $<1\mu\text{m}$ group (16%) and $1 - 2\mu\text{m}$ group (16%) compared to the $>2\mu\text{m}$ group (32%). Therefore, the bias between converted Dylos mass and Grimm mass varied by size group. Future studies examining particle size bins over a wider range of coarse particles ($> 2.5 \mu\text{m}$) would provide useful information for accurately converting LCPMS counts to mass concentration.

Keywords

Low-cost Sensors; Particulate Matter (PM); PM sensor calibration; Particle size

Introduction

Estimating particulate matter (PM) levels in the air is crucial in epidemiologic studies evaluating associations between this air pollutant and adverse health effects. Exposure to ambient $\text{PM}_{2.5}$ (aerodynamic size less than 2.5 micrometers) has been related to morbidity from respiratory health outcomes such as aggravation of asthma and chronic obstructive pulmonary disease (COPD) (Dominici et al. 2006; Sint et al. 2008), as well as cardiovascular health outcomes such as irregular heartbeat and strokes (Bourdrel et al., 2017; Dominici et al., 2006). $\text{PM}_{2.5}$ exposure is also a risk factor for premature death (deaths occurring before the average age of death, 75 years old) (Giannadaki et al. 2016; Liu et al. 2016) and mortality (Atkinson et al., 2014; Ostro et al., 2006; Zanobetti & Schwartz, 2009).

Low-cost particulate matter sensors (LCPMS) are handheld direct reading instruments (DRIs) equipped with low-cost sensors which specifically measure particulate matter (PM) concentrations in the air. The United States Environmental Protection Agency (US EPA) defines “low-cost sensors” in terms of cost as air monitoring devices that cost less than \$1,000 (USD) (Hall et al., 2014). In the last decade, LCPMS have been used to estimate exposure to $\text{PM}_{2.5}$ in the air within various indoor and outdoor settings (Han et al. 2017; Jiao et al. 2016; Jovasevic-Stojanovic et al. 2015; Steinle et al. 2015).

Currently, the gravimetric method is the standard for determining PM over time, usually 24 hrs in ambient environments and 8 hrs in workplace environments. Most federal reference method (FRM) monitors use the gravimetric method to measure the mass of particles deposited on a filter paper throughout sampling time for accurate and reliable measurements of $\text{PM}_{2.5}$. Local deployment of these FRM monitors at multiple locations is, however, limited by their cost and inability of real-time measurements. Although federal equivalence method (FEM) monitors such as the tapered element oscillating microbalance (TEOM) and beta attenuation monitor (BAM) are capable of continuous PM monitoring, these FEM monitors are still limited by cost, typically \$10,000 – \$20,000 (USD). Compared to the FEM monitors, the LCPMS provides real-time $\text{PM}_{2.5}$ measurements with relatively inexpensive costs (usually less than \$500 (USD)). Although other medium-cost DRIs such as nephelometers and photometers have been used for the $\text{PM}_{2.5}$ measurements, these DRIs

usually cost from \$3,000 – \$15,000 (USD). Given the fact, the LCPMS has the potential to monitor airborne PM in both indoor and outdoor settings by professional scientists and community instrument users (Jackson-Morris et al., 2016; Ramachandran et al., 2000; Rosen et al., 2015; Semple et al., 2015).

Measurements obtained from LCPMS are reported in number count per volume of air. To allow for comparisons with PM_{2.5} regulatory standards and other established PM_{2.5} monitoring methods (e.g., FRM or FEM), recent studies have compared PM_{2.5} measurements from the LCPMS to collocated PM_{2.5} measurements from a reference (e.g., FRM or FEM) or a research grade monitor. The correlations observed between the converted LCPMS measurements and measurements from reference or research grade monitors were from moderate to strong depending on the study ($R^2 = 0.44 - 0.98$) and the regression slopes used for the conversion of LCPMS measurements from particle counts to mass concentration also varied widely across these validation studies (Han et al., 2017; Jovasevic-Stojanovic et al., 2015; Northcross et al., 2013, Dacunto et al., 2015; Jiao et al., 2016). Variation in regression slopes has been partly attributed to some physical properties of the PM aerosols measured during calibration. Aerosol particle size, in particular, has been suggested to affect the LCPMS measurements. The intensity of light scattered by an aerosol is known to be dependent on the particle size distribution of an aerosol (Ramachandran et al., 2003). The effect of particle size distribution on submicrometer-sized particles (<1 μ m) increased slope with increasing particle size until 1 μ m. On the contrary, calibration slope was decreased for aerosols with particles greater than 2.5 μ m compared to aerosols with submicrometer sizes (Liu et al., 2017). This suggests that the LCPMS may require a different calibration curve when measuring aerosols composed of coarse particles (> 2.5 μ m) compared aerosols with fine particles (< 2.5 μ m). However, no analysis was conducted to determine the effect of aerosol particle size within the PM_{2.5} range (1– 2.5 μ m).

To accurately monitor PM_{2.5} with the LCPMS, it is essential to determine if different regression slopes will be required to calibrate the LCPMS with different aerodynamic particle sizes within the PM_{2.5} range. The main objective of this study was to assess the effect of aerosol particle size on the relationship between the Dylos DC1700 (LCPMS) PM_{2.5} measurements and GRIMM11R (a research grade instrument) PM_{2.5} measurements in a controlled laboratory chamber. A laboratory chamber study was selected where a single aerosol can be generated and measured as opposed to an ambient environment where complex mixtures of aerosols are usually present. Since the accuracy LCPMS measurements depend on the physical properties of the aerosols being measured, bias arising from measuring different aerosols was minimized.

Materials and Methods

Equipment

Dylos DC 1700 Air Quality Monitor (Dylos): The Dylos DC 1700 (Dylos Corporation, Riverside, CA) was selected as our LCPMS because of the availability of some research data for comparison from previous validation studies. The Dylos is a portable sensor that costs about \$425 USD. Light emitted by a photodiode is refracted by particles onto a sensor. The intensity of refracted light of the Dylos is converted to the number of particles by

two-particle size bins ($> 0.5 \mu\text{m}$ and $> 2.5 \mu\text{m}$). $\text{PM}_{2.5}$ concentrations measured by the Dylos are displayed in counts/ 0.01ft^3 for the two size bins. The Dylos counts can be converted to mass concentration using calibration equations that are usually developed from the linear regression of collocated measurements from the Dylos and the gravimetric or a research grade instrument in a laboratory chamber or a natural environment. With continued use over time in environments with extremely high PM concentrations, sensor measurement could be drifted (Collingwood et al., 2019). This drift or variation can be identified by checking baseline PM levels with zero air or in environments with low PM concentrations before and after use of the Dylos. Measurement variation may be due to degradation of the laser, degradation of the internal sensor, or changes in the flow rate of the fan. According to the manufacturer, cleaning and maintenance of the instrument can be performed by blowing compressed air into the openings in the rear of the Dylos. During the experiments in this study, we did not observe drifts or elevation of baseline from the Dylos.

Grimm Mini Laser Aerosol Spectrometer 11R (Grimm): The GRIMM 11R (Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany) was selected as the research grade monitor to which the Dylos $\text{PM}_{2.5}$ measurements were compared. The Grimm uses a laser spectrophotometric method to count the number of particles in 32 size bins ranging from $0.25 \mu\text{m} - 32 \mu\text{m}$. A density conversion factor developed by Grimm Aerosol Technik was used to convert the particle counts to mass concentration. This device costs about \$20,000 USD (~50 times the cost of the Dylos 1700).

Medline Disposable Nebulizer with Tee, Tubing, and Mouthpiece: A 10 mL Medline disposable nebulizer (Medline Industries Inc., Mundelein, IL) was used to disperse liquid sodium chloride placed in the cup into the air in aerosol form. The amount of aerosol dispersed was adjusted by varying the airflow from the pump. The size of the aerosol dispersed was adjusted by varying the concentration of the sodium chloride liquid placed in the cup. Prior studies also generated particles using an aerosol generator or a nebulizer with salt solutions of different concentrations (Li et al. 2020; Sousan et al. 2016).

HOBO Data Logger U12-012: The HOBO data logger (Onset Computer Corporation, Bourne, MA) was used to measure temperature and relative humidity with 1-min logging intervals.

SKC PCXR4 Universal Sample Pump: The PCXR4 sample pump (SKC Inc., Eighty-Four, PA) was used to provide pressurized air to the nebulizer. This pump was selected because of its ability to generate low volumes of air ($2 - 2.5 \text{ L/min}$), required in this study at a steady rate.

Experimental design

To assess the effect of particle size on the relationship between measurements taken by the Dylos DC1700 and the GRIMM11R, measurements of $\text{PM}_{2.5}$ aerosols with different median mass aerodynamic diameters (MMAD) were compared. One Dylos DC1700 and one GRIMM11R were placed within a chamber ($1\text{m} \times 1\text{m} \times 1\text{m}$) to measure $\text{PM}_{2.5}$ concentration. The HOBO data logger was used to record the average temperature and

relative humidity over one-minute periods within the chamber during sampling. Samples were collected from October 2019 through December 2019.

To generate aerosols, clean air ($PM_{2.5} < 1 \mu\text{g}/\text{m}^3$) using a PCXR4 Universal sample pump was pumped into the nebulizer cup through a Tygon tubing connected to the bottom of the nebulizer. Sodium chloride (NaCl) solution in the nebulizer cup was aerosolized by the airflow and dispersed into the chamber through the top of the nebulizer. Polydisperse sodium chloride crystals were used to make the NaCl solution in the cup. Sodium chloride crystals are commonly used as a test aerosol in previous studies (Li et al., 2020; Sousan et al., 2016). The aerosol was introduced into the chamber via an inlet on the side of the chamber and ventilated through an outlet at the top of the chamber. During sampling, the chamber was entirely sealed except the inlet to which the aerosol was introduced. Thus, possible contamination of other aerosols or dilution by clean air was prevented during the experiments. (Fig. 1)

By varying the concentration of the sodium chloride solution placed in the nebulizer, aerosols with MMAD of $< 1 \mu\text{m}$, $>1-2 \mu\text{m}$, and $>2 \mu\text{m}$ were generated. Through several pretests, it was determined that the concentrations of NaCl solution (in weight/volume%) required to generate aerosols with MMAD of $<1 \mu\text{m}$, $1-2 \mu\text{m}$ and $>2 \mu\text{m}$ were 0.01% NaCl, 0.5% NaCl and 40% NaCl, respectively. Real-time measurements of the particle size distribution of the aerosols within the chamber were monitored by the GRIMM during each experiment.

Sampling began one minute after the pump was turned on and ended when the particle size distribution returned to background levels. The Dylos 1700 and HOBO were set to log data throughout the sampling duration at 1 min intervals. The GRIMM 11R was set to a logging interval of 6 secs to allow for real-time monitoring of the $PM_{2.5}$ concentration changes within the chamber. Data analyses for all instruments were carried out using data averaged over one minute. For each particle size group, sampling was repeated until 300 one-minute data points were logged. (Table 1)

Data analysis

Data from the Dylos DC1700, the GRIMM 11-R, and the HOBO were downloaded to a laptop computer. The data from the Dylos DC 1700 included one-minute resolution particle counts for particles greater than $0.5 \mu\text{m}$ and particles greater than $2.5 \mu\text{m}$, along with the date and time these counts were collected. To obtain $PM_{2.5}$ particle number concentrations, the number counts for particles $> 2.5 \mu\text{m}$ were subtracted from the number counts for particles $> 0.5 \mu\text{m}$. The 1-min $PM_{2.5}$ mass data from the GRIMM 11R were downloaded. The 1-min relative humidity and temperature values were also downloaded. Data analysis was performed using STATA15 (StataCorp LLC., College Station, TX).

Mean $PM_{2.5}$ concentrations and standard deviations (SD), along with the minimum, maximum, and median concentrations, were summarized for the Dylos 1700 and the GRIMM 11R for the overall data and each MMAD group. After the data distributions for the Dylos 1700 and the GRIMM 11R were examined, log-transformed data were used for statistical analyses.

Second, the effect of particle size on the linear relationship between the Dylos 1700 and GRIMM 11R measurements was assessed. Product terms were added to the linear regression model (equation 1) to assess differences in slopes between the size groups at the significance level of $\alpha = 0.05$.

$$Y_i = \beta_0 + \beta_1 X_{i1} + \beta_2 X_2 + \beta_3 X_3 + \beta_4 (X_1 \times X_2) + \beta_5 (X_1 \times X_3) + \epsilon \quad (\text{eqn 1})$$

Where,

Y = natural log of the 1min $PM_{2.5}$ mass concentration measured by the Grimm

X_1 = natural log of the 1min $PM_{2.5-0.5}$ particle number concentration measured by the Dylos

X_2 = Binary variable coded as 1 for particle group size 1–2 μm and zero (0) for other particle size groups (<1 μm and >2 μm)

X_3 = Binary variable coded as 1 for particle group size >2 μm and zero (0) otherwise.

The particle size group <1 μm that coded as zero for both X_2 and X_3 is the reference group to which all the other particle size groups (<1 μm and >2 μm).

Third, the effect of particle size on the Dylos-Grimm relationship by different PM concentrations was evaluated after classifying the measurements into two different mass concentrations groups: Low PM (< 4920 particles/0.01ft³ or log-transformed (LN) Dylos count < 8.5) and High PM (>4920 particles/0.01ft³ or LN of Dylos count > 8.5).

Furthermore, the agreement (or bias) between the Dylos 1700 converted mass measurements and the GRIMM 11R measurements across particle size group was assessed using two methods:

1. Estimating the absolute relative error: Dylos 1700 number concentration measurements were converted to mass concentration measurements using the line equations from a linear regression of the data subdivided into particle size groups. Absolute relative error was calculated with equation 2 below.

$$\frac{|\text{Dylos } PM_{2.5} \text{ estimated mass concentration} - \text{Grimm } PM_{2.5} \text{ mass concentration}|}{\text{Grimm } PM_{2.5} \text{ mass concentration}} \quad (\text{eqn 2})$$

Where,

Dylos $PM_{2.5}$ = Converted $PM_{2.5}$ mass concentrations of the mean of 1 min Dylos measurements collected over a single sample duration

Grimm $PM_{2.5}$ = Mean of 1min $PM_{2.5}$ mass concentration measurements collected by the Grimm over a single sample duration

Analysis of variance (ANOVA) test was used to compare the difference between the mean absolute relative errors from the three particle size groups.

2. Bland-Altman analysis: The difference between each collocated Grimm 11R PM mass concentration and the estimated Dylos 1700 PM mass concentration (calculated from regression line equation for the overall data) was plotted

against the mean of the same Grimm 11R and Dylos 1700 measurements. The overall mean of the differences between Grimm 11R and Dylos 1700 mass concentrations was estimated with upper and lower confidence limits. A second Bland-Altman plot comparing the percent difference between the collocated Grimm 11R and Dylos 1700 measurements was also plotted. The percent difference was estimated by subtracting Dylos 1700 PM mass concentration from Grimm 11R mass concentration and dividing by the mean of the Grimm and Dylos measurements. The overall mean of all the percentage differences and confidence limits were examined. The Bland-Altman analysis was carried out using PM_{2.5} concentration data without log transformation. To determine the effect of particle size, a similar Bland-Altman analysis as described above was repeated after grouping the data into particle size groups (<1µm, 1 – 2µm, and >2µm).

Results and Discussion

Descriptive Statistics

The overall mean count (\pm SD (standard deviation)) for Dylos 1700 PM_{2.5} measurements was $10,963 \pm 9,370$ particles/0.01ft³ while the mean mass for Grimm 11R PM measurements was 45.6 ± 71.9 µg/m³. The overall mean temperature was 24.2 ± 0.1 °C and the overall mean relative humidity within the chamber was $21.8 \pm 18\%$. Medians and ranges for all parameters are summarized in Table 2.

The mean PM_{2.5} mass concentration from the Grimm 11R for particle size groups <1µm, 1–2µm, and > 2µm were 14.2 ± 10.3 µg/m³, 34.6 ± 27.1 µg/m³ and 88.2 ± 108.5 µg/m³, respectively (Table 3). The mean PM_{2.5} particle concentration from the Dylos 1700 for particle size groups <1µm, 1–2µm, and >2µm were $7,755 \pm 5,950$ particles/0.01ft³, $10,577 \pm 7,402$ particles/0.01ft³, and $14,556 \pm 12,265$ particles/0.01ft³, respectively. Mean temperature and relative humidity remained constant among three different particle size bins (Table 3).

Fig. 2 displays the mean 1-min PM_{2.5} concentration measured by the Dylos 1700 and Grimm 11R over the 300 mins of sampling. Generally, the Dylos and Grimm measurements follow a similar trend, although there is a slight time lag between the two instruments. This time lag can be explained by the difference in time from when each instrument is turned on to the logging of the first data point, which is longer in the Grimm. Within each particle size group, Grimm 11R PM_{2.5} mass (µg/m³) to Dylos 1700 PM_{2.5} count (500particles/0.01ft³) ratios increased with increasing particle size group. (<1 µm (ratio=1.34), 1 – 2 µm (ratio = 1.74) and >2 µm (ratio = 2.59)).

Effect of particle size on the linear relationship between the Dylos 1700 and Grimm 11R measurements

A simple linear regression was constructed for the overall data with the Grimm 11R measurements as the dependent variable and the Dylos 1700 measurements as the independent variable. The slope of the simple linear regression with the overall data was

0.93 ($R^2 = 0.89$). Fig. 3 shows that the slopes from the simple linear regressions by particle size group increased with the particle size bin. The slopes were 0.75 ($R^2 = 0.95$) for $<1\mu\text{m}$, 0.90 ($R^2 = 0.93$) $1\text{--}2\mu\text{m}$, and 1.03 ($R^2 = 0.94$) $>2\mu\text{m}$. These three slopes statistically differed from each other after testing for the significance of the coefficient of the interaction terms from Equation 1 ($p < 0.001$ [$1\mu\text{m}$ vs. $1\text{--}2\mu\text{m}$], $p < 0.001$ [$<1\mu\text{m}$ vs. $>2\mu\text{m}$], and $p < 0.001$ [$1\text{--}2\mu\text{m}$ vs. $>2\mu\text{m}$]).

Fig. 3 shows the association between Grimm and Dylos differed with increasing number concentration. The first cluster included data points with the natural log of Dylos count ≤ 8.5 (Dylos count ≤ 4920 particles/ 0.01ft^3), while the second cluster included data points with the natural log of Dylos count > 8.5 (Dylos count > 4920 particles/ 0.01ft^3). Table 4 summarizes that the slope was found to be statistically different (p -values < 0.001) for all three sizes between Dylos count $\leq 4,920$ (LN Dylos count ≤ 8.5) and Dylos count $> 4,920$ (Ln of Dylos count > 8.5).

Agreement between the estimated Dylos 1700 mass concentration and the Grimm 11R mass concentration

Absolute relative error: The mean absolute relative error (MARE) for the overall data was $22 \pm 18\%$. The MARE between measured Grimm 11R PM mass concentration and estimated Dylos 1700 PM mass concentration for aerosols with MMAD $<1\mu\text{m}$ ($16.5 \pm 16.8\%$) and MMAD $1\text{--}2\mu\text{m}$ ($16.4 \pm 15.5\%$) was similar. The MARE for MMAD $>2\mu\text{m}$ was $31.5 \pm 17.6\%$. A Bonferroni test confirmed that the MARE from the size group $>2\mu\text{m}$ was significantly greater than both the MARE from $<1\mu\text{m}$ and $1\text{--}2\mu\text{m}$ size group ($p=0.001$). The MAREs between the $<1\mu\text{m}$ and $1\text{--}2\mu\text{m}$ size groups were not statistically different ($p=1.0$).

Bland-Altman analysis: The overall mean bias between measured Grimm 11R mass concentration and the estimated Dylos 1700 mass concentration was $12.2\mu\text{g}/\text{m}^3$ with a standard deviation of $51.5\mu\text{g}/\text{m}^3$. The mean percentage bias was $-0.6 \pm 45.8\%$ (Table 5).

The Bland-Altman plots comparing the difference between the Grimm 11R and the Dylos 1700 by size group show that the mean absolute bias increases with increasing particle size. Similarly, the mean percentage bias also increased with an increase in particle size bins (Table 5).

Overall, co-located $\text{PM}_{2.5}$ measurements taken by the Dylos and the Grimm had a good correlation (coefficient of determination (R^2) for the linear regression of Grimm on Dylos measurements for the overall data was 0.89). This finding is similar to the R^2 obtained in prior studies comparing the Dylos monitor to other research grade instruments. For example, reported R^2 ranged from 0.7 – 0.99 between the Dylos DC1700 to the Tapered Element Oscillating Microbalance Filter Dynamics Measurement System (TEOM-FDMS) or the DustTrak 8520 in both outdoor environment and a laboratory chamber (Steinle et al. 2015, Northcross et al., 2013). The consistent results indicate that the moderate to good correlations between the Dylos and other research grade instruments are reproducible across different outdoor, indoor, and laboratory environments.

Given the regression slope of 0.93 with all data between Dylos and Grimm, Dylos count $PM_{2.5}$ measurements which are converted to mass concentration, do not seem to need much correction for bias. However, the relationship between the Dylos and the Grimm changes by three particle size groups ($<1\mu m$, $1-2\mu m$, and $>2\mu m$). We observed a statistical difference in the linear slopes across particle size groups. For aerosols with particle size less than $1\mu m$, the estimated $PM_{2.5}$ mass from Dylos overestimated 25 percent of the $PM_{2.5}$ concentration compared to the Grimm mass concentration (Slope = 0.75). Similarly, the Dylos overestimated 10 percent of $PM_{2.5}$ concentration compared to the Grimm for aerosols with the mean particle size of $1-2\mu m$ (Slope = 0.90) (*Note that regression slopes less than 1 indicate the Dylos overestimates PM concentrations against Grimm*). On the other hand, the Dylos for aerosols with mean particle size $>2\mu m$ slightly underestimated the Grimm (Slope = 1.03). The overestimation of Dylos can be explained by the Mie theory (Ramachandran et al., 2003). From Mie theory, for a given aerosol mass concentration, decreasing aerosol particle size would lead to increased intensity of scattered light (Ramachandran et al., 2003). An increased intensity of scattered light results in a higher response from the Dylos, leading to a larger $PM_{2.5}$ concentration measurement. The overestimation of Dylos for the smaller size of PM was also reported in urban ambient air in Houston, Texas. The ratio of Dylos PM measurements to Grimm 11R measurements was about 3:1 ratio when particle size was less than $1\mu m$ (Han et al., 2017). The findings suggest that particle size (especially size smaller than $2\mu m$) must be taken into consideration during the conversion and interpretation of measurements taken by the Dylos. For example, when using the Dylos to measure PM in locations where smaller sized particles are expected, such as close proximity to combustion sources or heavily trafficked roadways, a mean bias in converted Dylos mass can be predetermined from calibration of the Dylos with a reference instrument in the environment to be sampled. To provide improved correction factors for LCPMS, including Dylos, the association between LCPMS and a research grade instrument or a reference method should be examined under real-world environments with various combustion sources.

The effect of particle size on the Grimm-Dylos relationship was also changed by the ranges of PM counts. The slopes of between Dylos and Grimm measurements were less than 0.7 when PM concentration measured by the Dylos count was $4920 \text{ particles}/0.01\text{ft}^3$ (LN dylos < 8.5) compared to the slopes greater than 1 when Dylos count was $>4920 \text{ particles}/0.01\text{ft}^3$ (LN Dylos > 8.5) across all particle size groups. The results indicate that during the conversion and interpretation of measurements taken by the Dylos in areas with low particle count concentrations, such as in non-smoking homes or ambient settings with no significant sources of PM close by, Dylos measurement likely overestimates $PM_{2.5}$ mass concentration compared to research grade instruments. However, when particle count concentration is high with larger size particles ($> 2\mu m$) such as construction sites or suspended road dust, the Dylos measurements likely underestimate $PM_{2.5}$ mass concentrations than actual concentrations in those environments.

We observed that the absolute relative error for both particle size groups $<1\mu m$ (17%) and $1-2\mu m$ (16%) were significantly smaller than for the particle size group $>2\mu m$ (32%). A possible reason for the increased bias found in the larger particle size group ($>2\mu m$) is that the Dylos has a non-linear response to increasing PM concentration. The response of the Dylos was non-linear, becoming less responsive at higher PM concentration levels

due to the saturation of the sensor (Semple et al., 2012). The sensor saturation occurs at particle concentrations, after which further increase in signal to the instrument will result in little or no response for the instrument. The dynamic range of the Dylos has previously been reported at 10,000 particles/0.01ft³ (Han et al., 2017). Thus, when PM concentration approached the point of saturation, 10,000 particles/0.01ft³, the PM counts from Dylos reached a plateau resulting in increased bias. The Bland-Altman plot confirmed a positive trend in the magnitude of bias between the Grimm and the Dylos measurements as PM concentration increased.

A major strength of this study is that it was carried out in a laboratory chamber where confounding factors such as particle size, temperature, and relative humidity were controlled. Unlike studies carried out in ambient environments, the bias arising due to the presence of complex mixtures of particles and gases with different chemical and physical properties was avoided in the lab chamber. The aerosol measured in this study was composed of only NaCl aerosols; therefore, the generation of aerosols across different particle sizes was well controlled. Other atmospheric conditions such as temperature and relative humidity were also controlled. Relative humidity, in particular, has been suggested to affect the measurements from optical sensors. At high relative humidity (>60%), hygroscopic particles absorb water, increasing aerosol size and enhancing the intensity of light scattered by the particle (Chakrabarti et al., 2004; Han et al., 2017; Ramachandran et al., 2003).

Limitations of this study include the inability of the use of FRM or FEM for the comparison with Dylos. Although FEM gravimetric samplers are typically used as reference monitors when calibrating DRIs, including LCPMS, we did not use an FRM or FEM due to limited resources. However, the Grimm has been shown moderate to good correlations with FRM or FEM monitors in the past. For example, Grimm PM measurements had a good correlation with a filter dynamic measurement system (FDMS) monitor (R^2 range = 0.84 – 0.96) (Grimm & Eatough, 2009). The Grimm has been previously used to validate the quality of LCPMS measurements in several studies (Han et al., 2017; Holstius et al., 2014; Jovasevic-Stojanovic et al., 2015). In addition, we were unable to test inter-Dylos variability due to only one Dylos available in this study. However, previous studies demonstrated that the variations between multiple Dylos devices were less than 10 % (Klepeis et al., 2013; Wu et al., 2015). Another limitation includes that a wider range of particle size groups could not be examined in this study, especially monodisperse particles of larger size ranges (3 – 100 μ m). An advanced vibrating orifice aerosol generator (VOAG) can be used to generate particles over a wider range of particle sizes in future studies. Furthermore, while the conduct of this study inside a lab chamber environment allowed for the control of factors such as temperature and relative humidity, the association between the Dylos and Grimm in a real-world environment would be different from that in a chamber study. In natural environments, several factors such as the presence of complex mixtures of aerosols, gases, and meteorological conditions can modify the association between the Dylos and Grimm across the different particle size ranges as observed in this study. We expect the increase of variability and the decrease in the association between the Dylos and the Grimm measurements, in part, due to the presence of a complex mixture of particles and gases

under different meteorological conditions. Future studies also consider examining the effect of particle size on LCPMS measurements in various ambient air and indoor environments.

Conclusion

The effect of particle size distribution on the linear relationship between a low-cost PM sensor, Dyls DC1700, and a research grade instrument Grimm 11R was evaluated in this study. PM_{2.5} measurements obtained within a laboratory chamber were classified into three particle size groups (<1 µm, 1–2 µm, and > 2µm). A linear regression analysis was used to determine if the regression slopes between the Grimm and the Dyls varied by three particle size groups. The slopes between particle size groups statistically differed ($p < 0.001$). The slope within each particle size group was increased as PM_{2.5} concentration was increased. The biases between the Grimm and the Dyls were smaller in the <1µm and 1–2µm groups (16%) compared to the >2µm group (32%). Thus, both particle size and PM_{2.5} concentration are important factors that must be considered using LCPMS to measure PM_{2.5}. Given community instrument users of LCPMS may be unable to obtain information on the particle size of the calibration aerosol, correction factors for LCPMS should be provided by manufacturers to users at different ambient and indoor air settings.

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Research data supporting these data are available upon reasonable request.

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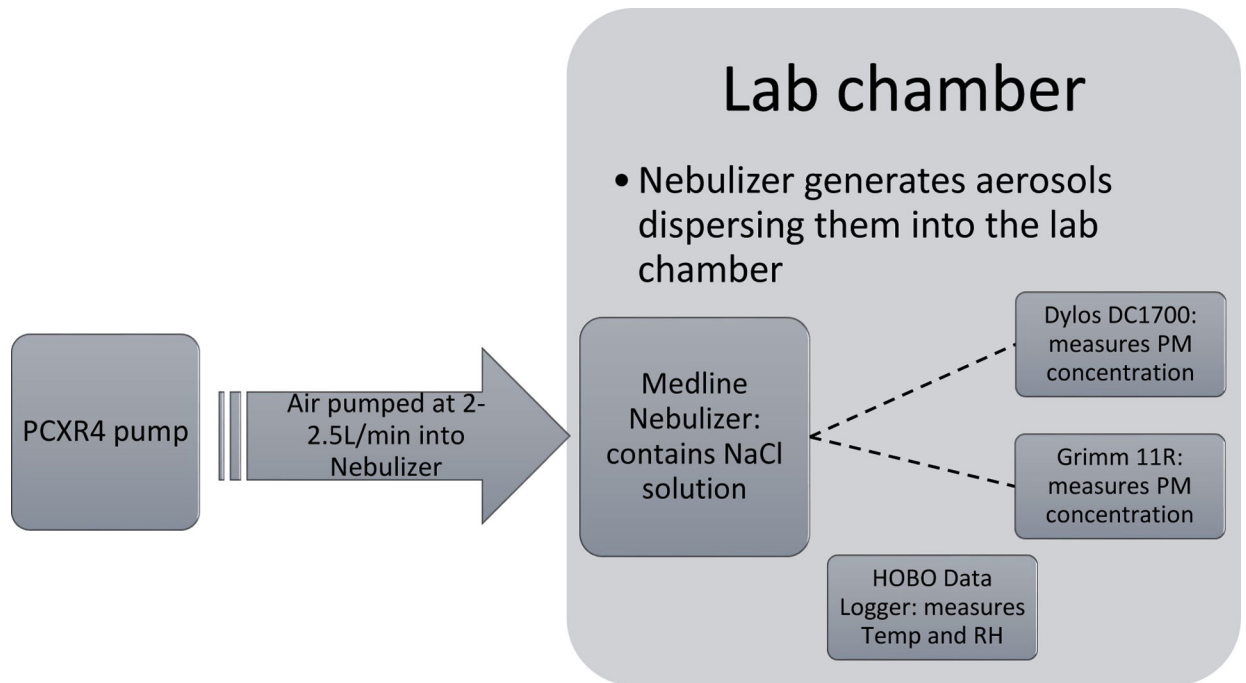


Fig. 1. Experimental design for the generation and measurement of PM_{2.5} aerosols with varying median mass aerodynamic diameters
 Definition: Flow chart generated with Microsoft word

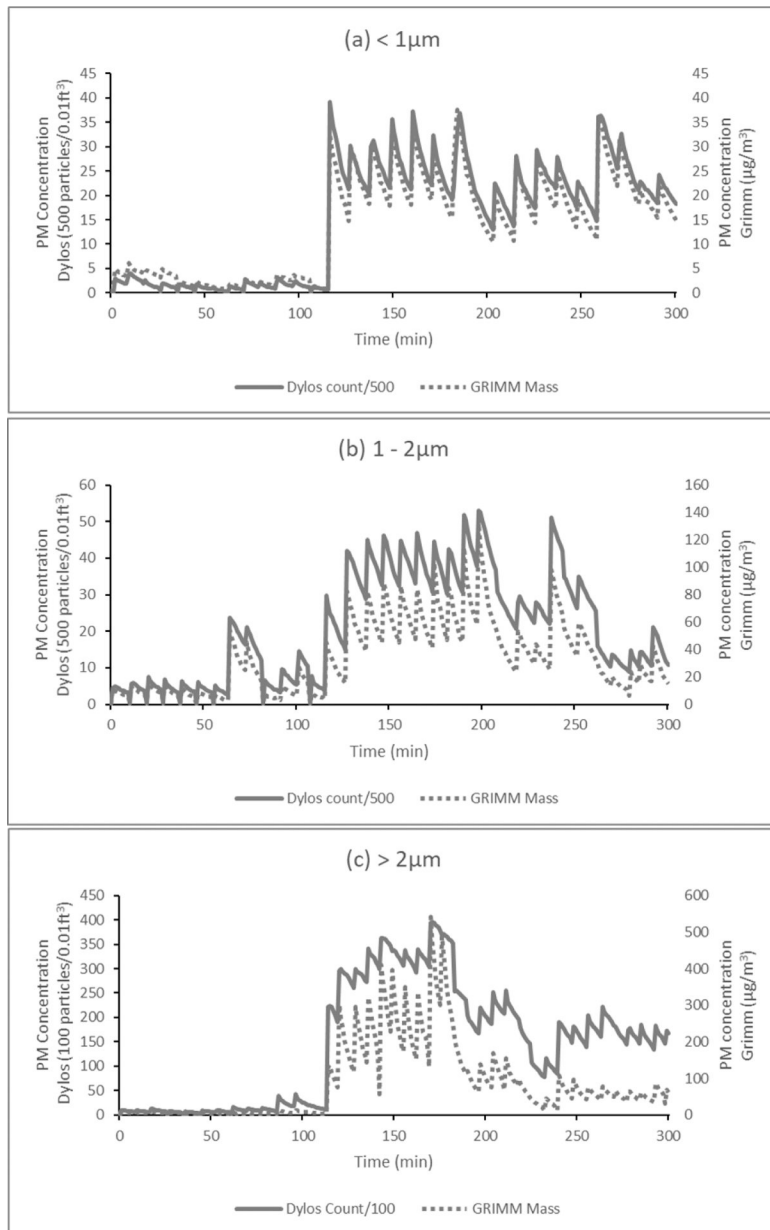


Fig. 2. Comparison of I-min average Dylos 1700 count to Grimm 11r mass by particle size group
Definition: Line plot generated with Microsoft excel

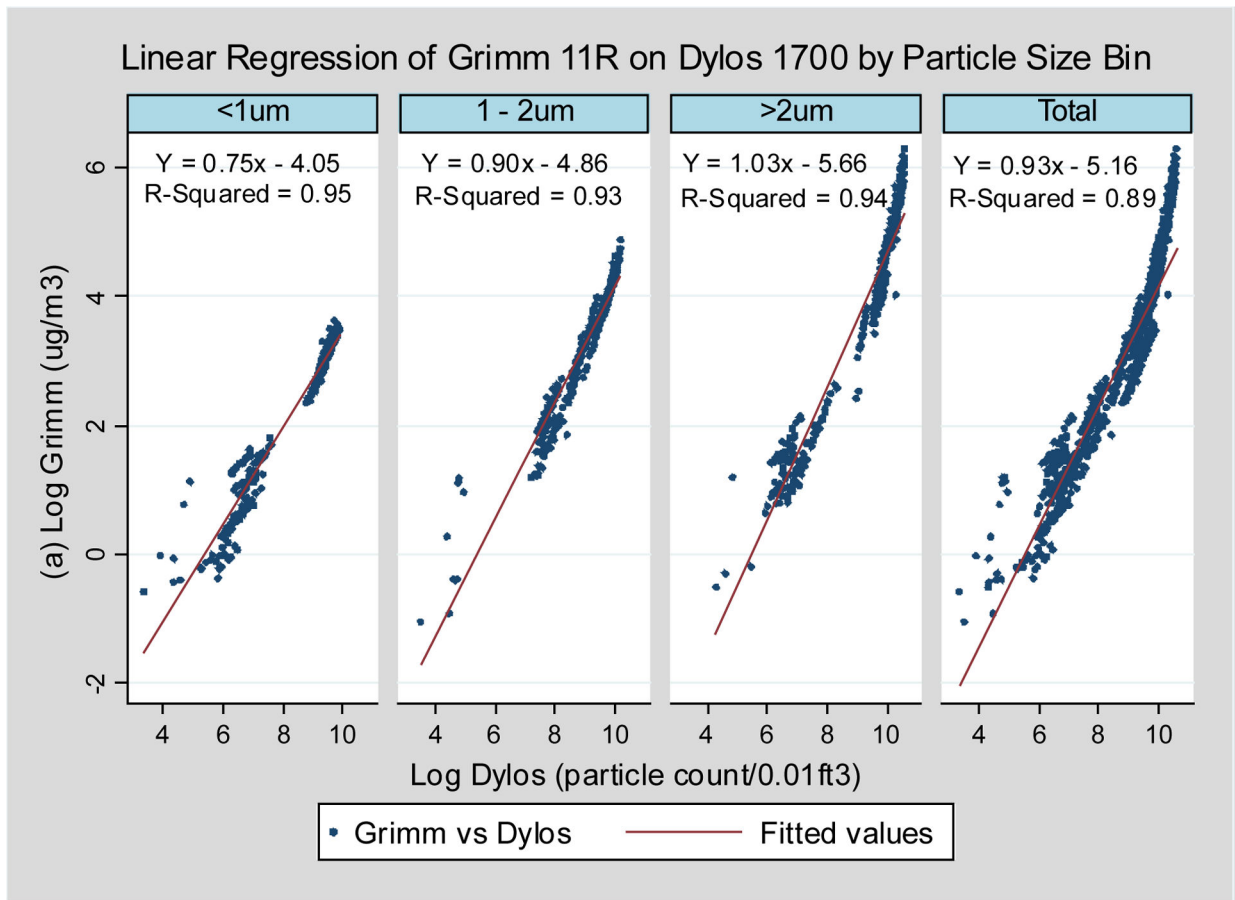


Fig. 3. Graph showing linear regression of Grimm measurements over Dylos measurements by particle size groups
 Definition: Scatter plot generated with STATA 15 statistical software
 Log grimm = Natural log Grimm PM_{2.5} measurements
 Log dylos = Natural log Dylos PM_{2.5} measurements.
 Each data point represents simultaneous 1-min PM_{2.5} measurements
 Sample size (n) = 900 for total data, n = 300 for each size group (<1µm, 1–2µm, and >2µm)

Table 1:

Experimental parameters for the generation of different aerosol sizes

Parameter	<1 μ m	1–2 μ m	>2 μ m
	(MMAD \pm SD) (0.48 μ m \pm 0.06)	(MMAD \pm SD) (1.12 μ m \pm 0.14)	(MMAD \pm SD) (2.75 μ m \pm 0.48)
NaCl Concentration	0.01%	0.5%	40%
Pump flow	2.5 L/min	2.5 L/min	2.0 L/min
1-min averaged PM data	n = 300	n = 300	n = 300

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Table 2:

Summary of data from Dyls 1700, Grimm 11R and HOBO.

Instrument	Measurement	a_n	Mean \pm SD	Median	Range
Grimm 11R	PM mass ($\mu\text{g}/\text{m}^3$)	900	45.66 ± 71.88	20.85	0.35 – 541.77
Dylos	PM number (particles/0.01ft ³)	900	10963 ± 9370	10570	28 – 39518
HOBO	Temp ($^{\circ}\text{C}$)	900	24.2 ± 0.1	24.4	21.3 – 25.5
HOBO	RH (%)	900	21.8 ± 13.8	15.0	15.0 – 51.9

^a $n = 900$ is composed of 3 size bins data: size $< 1\mu\text{m}$ ($n = 300$), size $1 - 2\mu\text{m}$ ($n = 300$), size $> 2\mu\text{m}$ ($n = 300$)

Table 3:

Summary of data from Dylus 1700, Grimm 11R and HOBO by particle size group

Instrument	Size group	n	Mean \pm SD	Median	Range
Grimm 11R ($\mu\text{g}/\text{m}^3$)	<1 μm	300	14.21 \pm 10.34	16.43	0.56 – 37.62
	1–2 μm	300	34.61 \pm 27.06	29.37	0.35 – 131.94
	>2 μm	300	88.15 \pm 108.49	52.02	0.60 – 541.77
Dylos (<i>particles/0.01ft³</i>)	<1 μm	300	77.5 \pm 59.5	98.0	0.28 – 195.9
	1–2 μm	300	10577 \pm 7402	9980	33 – 26500
	>2 μm	300	145.6 \pm 122.7	159.3	0.73 – 395.2
HOBO - Temp ($^{\circ}\text{C}$)	<1 μm	300	24.4 \pm 0.5	24.4	23.3 – 25.2
	1–2 μm	300	24.3 \pm 0.7	24.5	21.3 – 25.3
	>2 μm	300	23.9 \pm 1.4	24.4	21.4 – 25.5
HOBO - RH (%)	<1 μm	300	22.2 \pm 14.2	15.0	15.0 – 51.6
	1–2 μm	300	22.3 \pm 14.1	15.0	15.0 – 51.1
	>2 μm	300	21.0 \pm 13.1	15.0	15.0 – 51.9

Table 4:

Linear regression between Grimm and Dylos PM_{2.5} measurements by particle size group and PM concentration

Size group	Dylos Count 4,920 ^a	Dylos count > 4920 ^b
<1 μm	LN Grimm = (0.61 \times LN Dylos) – 3.14 (R ² =0.56)	LN Grimm = (1.20 \times LN Dylos) – 8.19 (R ² =0.94)
1–2 μm	LN Grimm = (0.61 \times LN Dylos) – 2.78 (R ² =0.75)	LN Grimm = (1.13 \times LN Dylos) – 6.95 (R ² =0.93)
>2 μm	LN Grimm = (0.64 \times LN Dylos) – 2.94 (R ² =0.65)	LN Grimm = (1.98 \times LN Dylos) – 15.13 (R ² =0.91)

^aGroup including Particle counts from the Dylos 4,920 or Natural Logarithm of the Dylos particle count 8.5

^bGroup including Particle counts from the Dylos > 4,920 or Natural Logarithm of the Dylos particle count > 8.5

Table 5:

Bland-Altman analysis comparing absolute difference and the percent difference between Grimm 11R and Dylos 1700 mass measurements

Size group	Difference Between Grimm and Dylos ($\mu\text{g}/\text{m}^3$)				Percent Difference Between Grimm and Dylos (%)			
	^a Mean	^b SD	^c LCL	^d UCL	^e Mean %	^a SD	^c LCL	^d UCL
<1 μm	-10.02	± 8.08	-25.86	5.81	-37.21	± 35.77	-107.33	32.90
1–2 μm	1.99	± 7.99	-13.66	17.65	0.03	± 26.75	-52.39	52.45
>2 μm	44.64	± 78.76	-109.73	199.01	35.30	± 40.91	-44.88	115.49
Total	12.20	± 51.54	-88.82	113.23	-0.62	± 45.80	-90.40	89.15

^aMean = $\Sigma (\text{Grimm} - \text{Dylos})/n$

^bSD = standard deviation,

^cLCL = 95th Lower Confidence Limit,

^dUCL = 95th Upper Confidence Limit

^eMean % = $\Sigma [(\text{Grimm} - \text{Dylos}) / \{(\text{Grimm} + \text{Dylos})/2\} * 100]/n$

Where Grimm = Grimm measurement, Dylos = collocated Dylos measurement, n = total number of measurements

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