

Ultrafine, fine, and black carbon particle concentrations in California child-care facilities

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Abstract

Although many U.S. children spend time in child care, little information exists on exposures to airborne particulate matter (PM) in this environment, even though PM may be associated with asthma and other respiratory illness, which is a key concern for young children. To address this data gap, we measured ultrafine particles (UFP), PM_{2.5}, PM₁₀, and black carbon in 40 California child-care facilities and examined associations with potential determinants. We also tested a low-cost optical particle measuring device (Dylos monitor). Median (interquartile range) concentrations for indoor UFP, gravimetric PM_{2.5}, real-time PM_{2.5}, gravimetric PM₁₀, and black carbon over the course of a child-care day were 14 000 (11 000–29 000) particles/cm³, 15 (9.6–21) µg/m³, 15 (11–23) µg/m³, 48 (33–73) µg/m³, and 0.43 (0.25–0.65) ng/m³, respectively. Indoor black carbon concentrations were inversely associated with air exchange rate (Spearman's rho = −.36) and positively associated with the sum of all Gaussian-adjusted traffic volume within a one-kilometer radius (Spearman's rho = .45) (*P*-values < .05). Finally, the Dylos may be a valid low-cost alternative to monitor PM levels indoors in future studies. Overall, results indicate the need for additional studies examining particle levels, potential health risks, and mitigation strategies in child-care facilities.

KEYWORDS

daycare, early childhood education, indoor air, predictors

1 | INTRODUCTION

Approximately 33% of United States children less than five years old regularly spend time in child-care or pre-school facilities, collectively known as early childhood education (ECE) facilities.¹ Although early life exposure assessments have focused on home environments, young children often spend a large portion of their waking hours at an ECE facility.² Previous studies in ECE facilities have characterized lead, pesticide, allergens, flame retardants, aldehydes, and phthalate exposures;^{3–8} however, little information exists on levels of particulate matter (PM) in child-care facilities, despite children being more susceptible than adults to the health effects of PM exposures.^{9–11}

Particulate matter is a complex mixture of small particles and liquid droplets. Constituents include inorganic compounds (such as nitrates

and sulfates), organic chemicals, metals, and soil or dust particles. Exposures of young children to PM_{2.5} (particles with an aerodynamic diameter ≤2.5 µm) and PM₁₀ (aerodynamic diameter ≤10 µm) have been shown to increase allergen sensitization, decrease lung function, and exacerbate pre-existing respiratory conditions like asthma.^{12–15} Ultrafine particle (UFP, aerodynamic diameter <0.1 µm) exposure to young children has been associated with decreased lung function¹⁶ and increased risk of respiratory hospital admissions.^{12–15,17,18} In children, black carbon exposure has been associated with lung oxidative stress,^{19–21} decreased lung function,²² and respiratory symptoms.^{23–25}

The costs to measure particles for health studies may be burdensome, forcing studies to limit the number of measurements or scope of work. Filter-based particle measurements require laboratory and technical training, but allow a more direct comparison to PM national

or state standards. Real-time samplers, which measure particles as a function of time, have costs that often exceed \$5000–\$10 000 USD. Previous research has shown that the Dylos monitor, a low-cost sampler (<\$300 USD), tracked well with higher cost samplers.^{26–28} However, it is unclear which method for converting Dylos measurements (count/unit volume) to the standard PM_{2.5} mass concentration (mass/unit volume) produces the best results. This is an important consideration for comparison with air quality standards and historical PM measurements, both typically expressed in mass concentrations.

In this study, we measured UFP, PM_{2.5}, PM₁₀, and black carbon at 40 ECE facilities in Northern California. We examined correlations between the particle concentration metrics and determinants of indoor and outdoor particle concentrations. We also assessed the performance of a low-cost optical particle sampler.

2 | MATERIALS AND METHODS

2.1 | Study population

The study population has been described previously.^{6,7} Briefly, we enrolled 40 ECE facilities from the California counties of Alameda (n = 20) and Monterey (n = 20), including 28 child-care centers and 12 home-based facilities to reflect the distribution of children in these types of facilities in California (60% and 40%, respectively).²⁹ All study protocols were approved by the University of California–Berkeley Committee for the Protection of Human Subjects, and informed written consent was obtained from each ECE facility program director or senior administrator.

2.2 | Study visits

Study field technicians typically visited a child-care facility twice. At the first visit, the technician administered a questionnaire and performed a site inspection focusing on the primary child-care room, cooking areas, and the bathroom. At the second visit, air samples were collected over the course of a school day (6–10 hours) when children were present. Indoor samples were collected in the main child-care room. The majority of outdoor measurements were collected in outdoor play areas. However, due to electrical power constraints or teacher preferences, some outdoor samples were collected in other areas adjacent to the facility. Site visits occurred from May 2010 to May 2011.

2.3 | Gravimetric PM sampling and analysis

Gravimetric PM_{2.5} and PM₁₀ air samples were collected using SKC[®] Personal Environmental Monitors (PEMs) (SKC Inc., Eighty Four, PA, USA) onto 37-mm Teflon filters. Due to cost constraints, PEMs with required flow rates of two and four liters per minute (LPM) were used. The PEMs with a four LPM flow rate were typically used indoors (n = 31) and PEMs with a two LPM flow rate were typically used outdoors (n = 12). Indoor air samples were pulled through filters using a fan-cooled, single rotary vane pump installed in a stainless steel box lined with foil-faced fiberglass sound insulation to reduce noise. To

Practical Implications

- This is the first study to report on a wide array of particle exposures in U.S. early childhood education (ECE) environments. Levels of particles may pose a health risk to children, as we found indoor PM_{2.5} and PM₁₀ concentrations collected during the school day exceeded 24-hour ambient air quality standards in some facilities. Further, correlations between particle metrics measured in this study indicate that children are receiving co-exposures to the various sizes and composition of particles; therefore, subpopulations of children may be at increased risk of the health effects of early life particulate exposure. Due to the large number of children spending a significant amount of time in ECE facilities, our research highlights that exposure mitigation may be warranted.

eliminate emissions, the pump's exhaust system included a muffler and a HEPA and carbon filter. Outdoor air samples were collected using SKC AirChek 2000 pumps (SKC Inc.).

Flow rates were set per the manufacturer's recommendations, and samples were integrated over the sampling period. Flow from the rotary vane pump was controlled via taper-tube flowmeters (Key Instruments, part # 10510 and 10710). Calibration curves were determined for each flowmeter with filters in-line using a Gilibrator[®] airflow calibrator (Sensidyne, St. Petersburg, FL, USA). Calibration curves were checked at the end of the sampling campaign and were consistent with results prior to sampling. Flow from the AirCheks was calibrated before and after each use with a Gilibrator[®] airflow calibrator.

The gravimetric PM_{2.5} and PM₁₀ particle mass concentrations were determined following EPA Method IP-10A.³⁰ Each Teflon filter used for mass analysis was weighed on two separate occasions before deployment and after sampling using a Sartorius SE-2F balance. Filters were equilibrated for a minimum of 24 hours at 21 ± 3°C and 30%–40% relative humidity for at least one weighing before and one weighing after sampling. A 100 µg certified standard weight was weighed with each group of sample filters to confirm consistent operation of the balance. A method detection limit (MDL) of 14.4 µg was calculated by computing three times the standard deviation of blank filters. Reported data were blank corrected by subtracting the mean blank mass (2.3 µg) from weighed particle masses. During the pilot stage of the project (facilities 1–5), filter contamination occurred due to the gaskets in some of the PEM bodies failing and shedding mass onto the filters during the loading and unloading process. Upon finding this flaw, all PEM bodies were reconditioned and confirmed that weight change of the filters during loading and unloading was within acceptable limits (i.e., <3 µg change between measurements). Therefore, only 35 indoor gravimetric PM samples are reported in this analysis. To assess quality control, eighteen Teflon filter field blanks were weighed prior to sampling, taken into the field by a technician without exposing the PEM, and then returned to LBNL for post-weighing. The field blanks had a

mean (SD) mass of 2.3 (4.8) μg . Four duplicate measurements were collected indoors for each $\text{PM}_{2.5}$ and PM_{10} for comparison. For $\text{PM}_{2.5}$, one pair of duplicates were both below the MDL, one pair was both below and above the MDL (although the one detected was only 0.9 μg above the MDL), and two were both above the MDL. For the two duplicate measurements above the MDL, the relative percent differences (RPDs) were 48% and 54%. The larger RPDs for $\text{PM}_{2.5}$ are likely due to the measurements being below/close to the MDL. The difference in PEMs (one a 4 LPM PEM, another a 2 LPM PEM) between duplicate measurements may have also added to the variability. For PM_{10} , all four of the duplicates were above the MDL and the RPD ranged between 3.9% and 15%. Please see the Supplemental Information (SI) for additional QA/QC information for these instruments.

2.4 | Real-time measurements

We used real-time instruments to measure concentrations of UFPs, $\text{PM}_{2.5}$, black carbon, and carbon dioxide (CO_2) during a full school day. UFPs were measured using two TSI Inc. 3781 water condensation particle counters (CPC). The TSI 3781 detects (D_{50} —detection of 50% of particles) total particle number concentration in the size range of $>3 \mu\text{m}$ down to 6 nm and concentration of 0 to 5×10^5 particles/ cm^3 . UFP counts were collected indoors in 39 facilities (in one instance the CPC did not record the data properly) and outdoors in 28 facilities. Quality control included collection of co-located UFP measurements, conducted at two time points. The measurements were highly correlated (Pearson's $R^2 = 0.98$ and 0.99 , respectively) with a median RPD of 2.0% at each time point and a median absolute deviation (MAD) of 1.6% and 8.8%, respectively (Fig. S1).

Real-time fine-particulate matter was measured with a TSI DustTrak 8520 (DT1) and 8530 (DT2). The DustTrak 8520 has an aerosol concentration range of 0.001 to 100 mg/m^3 , while the TSI 8530 has a concentration range of 0.001 to 400 mg/m^3 . Both DustTraks were calibrated prior to field deployment by TSI, used 2.5 μm size selectors, and were zeroed prior to each use. Side-by-side measurements between DT1 and DT2 before the sampling campaign showed a strong correlation ($R^2 = 0.80$). However, there was a consistent bias with DT1 on average 7 $\mu\text{g}/\text{m}^3$ lower than DT2; the MAD (RPD) was 13% (2.0%). Side-by-side comparisons of DT1 and DT2 after the sampling campaign also correlated well ($R^2 = 0.95$), but a bias persisted between the machines as DT1 was approximately 5 $\mu\text{g}/\text{m}^3$ lower than DT2 throughout the sampling period. The post-sampling MAD (RPD) was 24% (3.2%) (Fig. S2). Overall, QA procedures for the DustTraks before and after sampling indicate strong correlations between DT1 and DT2 $\text{PM}_{2.5}$ measurements, but the DT2 levels averaged 25% higher. Due to these findings, the DT2 measurements were adjusted downward with a correction offset of $-6 \mu\text{g}/\text{m}^3$ to be comparable to the DT1 results.

Magee Scientific's MicroAeth AE51s were used to measure real-time black carbon concentrations within a range of 0–1 mg/m^3 with a resolution of 1 ng/m^3 . Indoor real-time black carbon concentrations were measured at all ECE facilities ($n = 40$). Side-by-side comparisons between the two MicroAeths were performed in six facilities. The R^2 between the duplicate minute-by-minute MicroAeth measurements was 0.49 and the MAD (RPD) was 31% (30%) (Fig. S3).

We used two Dylos DC1100 Pro instruments that measure particles with an aerodynamic diameter greater than or equal to 2.5 μm ($\text{PM}_{\geq 2.5}$) and particles with an aerodynamic diameter greater than or equal to 0.5 μm ($\text{PM}_{\geq 0.5}$). The Dylos uses a laser light scattering method for size selection. The Dylos gives hourly average concentrations (particles/ ft^3) and a rolling minute-by-minute particle concentration for the last 60 minutes of sample time. Side-by-side comparisons between two Dylos were performed in four facilities. The R^2 between the duplicate minute-by-minute Dylos measurements was 0.92 for the small particles and 0.74 for the large particles. The MAD (RPD) for the small and large particles was 8.1% (4.2%) and 10% (8.8%), respectively (Fig. S4).

2.5 | Air exchange and ventilation rates

Our method for estimating air exchange rate (AER) has been described previously.^{6,8} Briefly, we estimated AER using a continuous indoor CO_2 mass balance model, supplemented with a single CO_2 tracer-gas release.^{31,32} The mass balance model estimated the AER by minimizing the sum of mean squared error between measured (via QTrak 8554 from TSI) and modeled CO_2 concentrations taking into account room occupancy and conditions (e.g., windows/doors opening). For the CO_2 tracer-gas release, we released medical-grade CO_2 (Praxair, part number CD M-10, pharmacopeia grade) when children were not present to elevate CO_2 levels to approximately 2500 ppm. By combining both of these methods in each ECE facility, we were able to model the air exchange rate under normal room conditions, plus match our air exchange models to an enriched CO_2 environment to ensure our model fit multiple scenarios. In addition to AER, we calculated the ventilation rate for each facility by multiplying the AER with the room volume and dividing by the weighted average of room occupancy (m^3/hour per person).

2.6 | Traffic metrics

We obtained three traffic statistics within a one kilometer radius buffer for each ECE facility from the California Environmental Health Tracking Program traffic linkage service on July 19th, 2011.³³ The following traffic summary statistics were abstracted: sum of all length-adjusted traffic volumes (ΣLATV), which is the sum of the number of vehicles per hour multiplied by the total length of road within the buffer (vehicle-km/hour); sum of all Gaussian-adjusted traffic volumes (ΣGATV), which is the distance weighted sum of traffic volumes (vehicles/day) assuming a Gaussian dispersion of airborne exhaust pollution from the traffic segment and weighted by a 500 m radius (half 1-km total radius);³⁴ and length-adjusted traffic volume of the highest segment (LATV-HS) which calculated the LATV of the heaviest used road within the buffer (vehicle-km/hour).

2.7 | Data analyses

2.7.1 | Descriptive analysis of particle data

When duplicate measurements were collected for a facility, the average concentration between the two measurements by time point

(real-time sampling) or facility (gravimetric sampling) was calculated and reported. In the rare circumstance that the DustTrak recorded a zero value, the value was replaced with the lowest concentration resolution ($1 \mu\text{g}/\text{m}^3$) divided by $\sqrt{2}$.³⁵ For gravimetric $\text{PM}_{2.5}$ and PM_{10} values below the MDL, we used the weighed mass below the MDL to calculate the arithmetic and geometric mean and standard deviations.

For real-time UFP, $\text{PM}_{2.5}$, PM_{10} , and black carbon, we used generalized additive models to show the changes in concentrations over the course of the day. For real-time measurements, we calculated the overall mean values of particle measurements in each facility. We then calculated both arithmetic and geometric means and standard deviations, along with selected quantiles of the distribution. We computed Spearman's rank correlation coefficients to test the correlation between particle metrics, traffic metrics, and air exchange rate. We evaluated differences in particle concentrations by ECE location (Alameda vs Monterey County) and presence of combustion appliances in or adjacent to the sampling room using unpaired Wilcoxon-Mann-Whitney tests.

Indoor-to-outdoor (I/O) air concentration ratios were computed by dividing the average indoor air concentration by the outdoor air concentration for each facility with paired measurements. We used the paired Wilcoxon-Mann-Whitney test to evaluate differences in indoor and outdoor particle concentrations.

2.7.2 | Comparison of real-time Dylos and DustTrak particle measurements

Minute-by-minute particle concentrations were measured by both the Dylos and DustTrak indoors in 32 facilities and outdoors in seven facilities. To compare the Dylos measurements with $\text{PM}_{2.5}$ measured by the DustTraks, we subtracted the difference between $\text{PM}_{2.5}$ and $\text{PM}_{2.0.5}$ count concentrations (herein $\text{PM}_{0.5-2.5}$).^{27,28} We then fit a linear regression of all paired minute-by-minute Dylos $\text{PM}_{0.5-2.5}$ count concentrations against the DustTrak mass concentrations. We also evaluated two approaches to convert particle count concentration (particles per volume air – $\text{conc}_{\text{count}}$) to particle mass concentration (mass of particles per volume of air). The first, developed by Semple et al²⁷ and Steinle et al,²⁸ were developed for indoor,²⁷ outdoor

rural,²⁸ and outdoor urban locations.²⁸ We used the Steinle et al²⁸ outdoor rural equation for all Monterey ECE outdoor measurements and the outdoor urban equation for Alameda county ECE outdoor measurements. Northcross et al²⁶ used the properties of a sphere (assuming an average aerodynamic radius (r_p) of the particles to be $0.75 \mu\text{m}$) and particle density (ρ_p , assumed to be $1.0 \text{g}/\text{cm}^3$) to calculate mass concentrations. All four of the equations used are presented in the SI (Table S1).

2.7.3 | Comparison with ambient air quality standard concentrations

We compared our measured gravimetric $\text{PM}_{2.5}$ and PM_{10} concentrations with the levels of the California Ambient Air Quality Standards (CAAQS) for annual ($\text{PM}_{2.5} = 12 \mu\text{g}/\text{m}^3$ and $\text{PM}_{10} = 20 \mu\text{g}/\text{m}^3$) and 24-hour exposures ($\text{PM}_{10} = 50 \mu\text{g}/\text{m}^3$).³⁶ As there is no 24-hour CAAQS for $\text{PM}_{2.5}$, we compared our $\text{PM}_{2.5}$ values to the level of the 24-hour National Ambient Air Quality Standard of $35 \mu\text{g}/\text{m}^3$.³⁷

All statistical analyses were performed with R Version 3.1.3.³⁸

3 | RESULTS

3.1 | ECE facility characteristics

Descriptions of the facilities have been previously published.⁶⁻⁸ Briefly, 1764 children were served by the programs, and the average attendance was 44 children per facility, with a maximum attendance of 200 children. Twenty-six (65%) were in residential neighborhoods, eight (20%) were in commercial areas, five (13%) were adjacent to agricultural fields, and one (3%) was in a rural/ranch area. Half were in buildings constructed after 1970, with the oldest structure built in 1903 and the most recent in 2008. Heating systems were on average 16 years old (range = 1-80 years). The average (SD) air exchange and ventilation rate were 2.0 (1.4) per hour and 40 (32) m^3 per hour per person during air sampling, respectively. Natural ventilation (i.e., opening windows) was used in most of the facilities (91%) due to the moderate climate in Northern California.

TABLE 1 Summary of particle concentrations in indoor air

Metric ^a	n	%Det ^b	Mean	SD ^b	GM ^b	GSD ^b	Min	25 th %	50 th %	75 th %	Max
Ultrafine particles per cm^3	39	-	22 000	20 000	16 000	2.4	1500	11 000	14 000	29 000	75 000
Gravimetric $\text{PM}_{2.5}$ ($\mu\text{g}/\text{m}^3$)	35	80	18	11	15	1.8	<MDL	9.6	15	21	54
Real-time $\text{PM}_{2.5}$ ($\mu\text{g}/\text{m}^3$)	40	-	19	16	16	1.9	5.2	11	15	23	89
Gravimetric PM_{10} ($\mu\text{g}/\text{m}^3$)	35	100	55	32	47	1.8	14	33	48	73	170
Black carbon ($\mu\text{g}/\text{m}^3$)	40	-	0.57	0.52	0.38	2.8	0.16	0.25	0.43	0.65	2.5

^aThe average concentration over the child-care day for each facility is presented here.

^b% Det., percent above method detection limit; SD, standard deviation; GM, geometric mean; GSD, geometric standard deviation. For gravimetric PM samples, mass below the detection limit was used to calculate the mean, SD, GM, and GSD.

TABLE 2 Summary of particle concentrations in outdoor air

Metric ^a	n	%Det ^b	Mean	SD ^b	GM ^b	GSD ^b	Min	25 th %	50 th %	75 th %	Max
Ultrafine particles per cm ³	28	-	17 000	11 000	13 000	2.1	1300	9900	14 000	20 000	45 000
Gravimetric PM _{2.5} (µg/m ³)	12	83	18	9.3	9.8	3.3	<MDL	8.2	13	21	37
Real-time PM _{2.5} (µg/m ³)	31	-	24	28	17	2.3	2.4	12	17	24	140
Gravimetric PM ₁₀ (µg/m ³)	12	50	40	27	7.2	8.1	<MDL	<MDL	13	43	94

^aThe average concentration over the child-care day for each facility is presented here.

^b% Det., percent above method detection limit; SD, standard deviation; GM, geometric mean; GSD, geometric standard deviation. For gravimetric PM samples, mass below the detection limit was used to calculate the mean, SD, GM, and GSD.

3.2 | Particle measurements and correlations

The median (interquartile range (IQR)) of the average ultrafine particle concentration, monitored by a CPC, over the sampling period was 14 000 (11 000-29 000) particles/cm³ (Table 1). Gravimetric PM_{2.5} and PM₁₀ mass was above the detection limit in 80% and 100% of the indoor samples (n = 35), respectively, and 83% and 50% of the outdoor samples, respectively. The distributions of the indoor gravimetric (n = 35) and real-time PM_{2.5} (n = 40) were similar with median (IQR) concentrations of 15 (9.6-21) and 15 (11-23) µg/m³, respectively. In 47 side-by-side indoor and outdoor measurements, we found gravimetric measurements to be typically lower than average real-time PM_{2.5}, with the median (IQR) ratio between the gravimetric and real-time measurements to be 0.86 (0.72-1.2). The distribution of real-time PM_{2.5} was higher outdoors compared to indoors (median outdoor (IQR) = 17 (12-24) µg/m³) (Table 2).

Only gravimetric PM₁₀ showed a tendency to be higher indoors as compared to outdoors (median I/O ratio = 1.8, *P*-value = .08) (Table 3). The other metrics measured both indoors and outdoors (UFP, gravimetric PM_{2.5}, and real-time PM_{2.5}) were not significantly different (*P*-value >.1; median I/O ratios = 1.2, 1.0, and 0.9, respectively). However, in one extreme instance, the indoor ultrafine concentration was, on average, 38 times higher indoors than outdoors, likely due to simultaneous cooking with an unvented gas stove (Fig. S5).

We found multiple significant correlations between the particle metrics (Table 4). For example, indoor black carbon was positively

associated with five of eight other particle metrics including indoor real-time PM_{2.5} (rho = .83), outdoor gravimetric PM₁₀ (rho = .76), outdoor real-time PM_{2.5} (rho = .74), indoor gravimetric PM_{2.5} (rho = .52), and indoor gravimetric PM₁₀ (rho = .45) (*P*-values <.01).

Indoor UFP tended to increase in the morning and decrease in the late afternoon (Figure 1), likely due to use of combustion appliances for morning food preparation and traffic. Outdoor UFP tended to peak in the morning and early afternoon. Both indoor and outdoor PM_{2.5} increased in the morning with similar slopes, but indoor PM_{2.5} had a cyclical pattern, while outdoor PM_{2.5} stayed relatively constant during the mid-morning and early afternoon. However, outdoor PM_{2.5} typically increased in the late afternoon, potentially due to increased traffic. Similar to indoor PM_{2.5}, black carbon tended to increase in the morning, decrease in the mid-morning, then increase in the late afternoon—possibly tracking morning/evening traffic patterns.

3.3 | Determinants of particle concentrations

There were wide variations in traffic density reflecting the urban and rural locations (Table S2). Indoor black carbon and outdoor real-time PM_{2.5} were positively correlated with ΣGATV (rho = .45 and rho = .62, respectively; *P*-values <.01); however, the other indoor or outdoor particle metrics were not associated with ΣLATV, ΣGATV, or LATV-HS (Table S3). While all particle concentrations except indoor UFP were inversely associated with AER, only black carbon was statistically significant (rho = -.36, *P*-value <.05).

TABLE 3 Summary of particle indoor/outdoor (I/O) ratios

Metric ^a	n	GM ^b	GSD ^b	Min	25 th %	50 th %	75 th %	Max	<i>P</i> -value ^c
Ultrafine	27	1.2	2.8	0.3	0.7	1.2	1.9	37.9	.31
Gravimetric PM _{2.5}	12	1.0	1.5	0.6	0.8	1.0	1.2	2.6	.97
Real-time PM _{2.5}	31	0.9	2.1	0.2	0.6	0.9	1.1	7.6	.14
Gravimetric PM ₁₀	12	1.6	1.9	0.6	1.1	1.8	2.1	4.8	.08

^aI/O ratios calculated with average real-time concentrations, when applicable.

^bGM, geometric mean; GSD, geometric standard deviation. For gravimetric PM samples, mass below the detection limit was used to calculate the GM and GSD.

^c*P*-values from paired Wilcoxon-Mann-Whitney rank sum tests evaluating the differences in indoor and outdoor particle concentrations.

TABLE 4 Spearman's rho correlation matrix comparing different particle matter concentration/count metrics^{a,b}

Metric	Indoor ultrafine	Indoor gravimetric PM _{2.5}	Indoor gravimetric PM ₁₀	Indoor real-time PM _{2.5}	Indoor gravimetric PM ₁₀	Indoor black carbon	Outdoor ultrafine	Outdoor gravimetric PM _{2.5}	Outdoor gravimetric real-time PM _{2.5}
Indoor ultrafine	1	-	-	-	-	-	-	-	-
Indoor gravimetric PM _{2.5}	0.25 (34)	1	-	-	-	-	-	-	-
Indoor real-time PM _{2.5}	0.16 (39)	0.71** (35)	1	-	-	-	-	-	-
Indoor gravimetric PM ₁₀	0.07 (34)	0.79** (35)	0.67** (35)	1	-	-	-	-	-
Indoor black carbon	0.10 (39)	0.52** (35)	0.83** (40)	0.45** (35)	1	-	-	-	-
Outdoor ultrafine	0.35 (27)	-0.15 (28)	0.04 (28)	0.04 (28)	-0.45* (28)	0.30 (28)	1	-	-
Outdoor gravimetric PM _{2.5}	-0.30 (12)	0.80** (12)	0.48 (12)	0.48 (12)	0.54 (12)	0.45 (12)	0.04 (12)	1	-
Outdoor real-time PM _{2.5}	-0.38* (30)	0.31 (31)	0.67** (31)	0.67** (31)	0.39* (31)	0.74** (31)	0.04 (28)	0.49 (12)	1
Outdoor gravimetric PM ₁₀	-0.12 (12)	0.43 (12)	0.69* (12)	0.69* (12)	0.57 (12)	0.76** (12)	0.38 (12)	0.61* (12)	0.76** (12)

^aNumbers in parenthesis are number of paired particle measurements used in correlation calculation.

^b*, Spearman P -value < .05; **, Spearman P -value < .01.

Indoor and outdoor particle concentrations were typically higher in facilities located in Alameda compared to Monterey County (Table S4). For example, indoor black carbon concentrations were significantly higher in urban Alameda County (median (IQR) = 0.57 (0.35-1.1) $\mu\text{g}/\text{m}^3$) compared to less developed Monterey County (median (IQR) = 0.33 (0.21-0.47) $\mu\text{g}/\text{m}^3$) (P -value = .03). Many of the Alameda County ECE facilities were also located near the Interstate 80-880 freeways and/or the Oakland Port, which carry heavy diesel truck traffic.³⁹

The presence of a combustion appliance (typically natural gas stove, dryer, or furnace) in or adjacent to the room where air samples were collected was moderately associated with higher UFP (Table S5, P -value = .07). For example, in the 13 facilities with a combustion appliance present, the median (IQR) was 28 000 (12 000-48 000) particles/ cm^3 , vs 12 000 (9700-18 000) particles/ cm^3 in facilities without a nearby combustion source. No other indoor particle metric was associated with the presence of combustion source (P -values > .1). As noted, we observed large, discernable spikes in UFP concentrations when gas appliances were used (Fig. S5).

3.4 | Comparison of real-time particle sample methods

We observed good agreement between the minute-by-minute Dylos PM_{0.5-2.5} and DustTrak's PM_{2.5} ($R^2 = 0.82$, Fig. S6). The linear model relating the Dylos PM_{0.5-2.5} count measurements per ft^3 to DustTrak PM_{2.5} concentrations ($\mu\text{g}/\text{m}^3$) was the following:

$$\text{DustTrak PM}_{2.5} = 0.52 + 9.4e - 5 * \text{Dylos PM}_{0.5-2.5} \quad (1)$$

The root mean squared error (RMSE) between the predicted and observed DustTrak PM_{2.5} using Equation 1 was 6.5 $\mu\text{g}/\text{m}^3$. When we removed the outlier concentrations in the upper right of Fig. S6, the linear model was the following:

$$\text{DustTrak PM}_{2.5} = 3.1 + 7.6e - 5 * \text{Dylos PM}_{0.5-2.5} \quad (2)$$

Without the outliers, both the RMSE and R^2 decreased to 5.9 and 0.65, respectively. When we compared the two methods proposed to convert Dylos count concentrations to mass concentrations, we found that using the method proposed by Northcross et al²⁶ matched our DustTrak measurements better than using the methods proposed by Steinle et al²⁸ (Figure 2). The RMSE and R^2 were 6.7 $\mu\text{g}/\text{m}^3$ and 0.83, respectively, using the Northcross et al²⁶ method, whereas the RMSE and R^2 were 13 $\mu\text{g}/\text{m}^3$ and 0.79, respectively, using the Steinle et al²⁸ method. When we removed the outliers in the top right of Figure 2, Northcross et al's method (RMSE = 6.6 $\mu\text{g}/\text{m}^3$ and $R^2 = 0.65$) still matched better Steinle et al's method (RMSE = 11 $\mu\text{g}/\text{m}^3$ and $R^2 = 0.52$).

3.5 | Comparison with ambient air quality standard concentrations

When comparing our indoor gravimetric PM concentrations to the levels of the CAAQS and NAAQS (24-hour PM_{2.5} only), we found that the indoor PM_{2.5} concentrations exceeded the levels of the annual

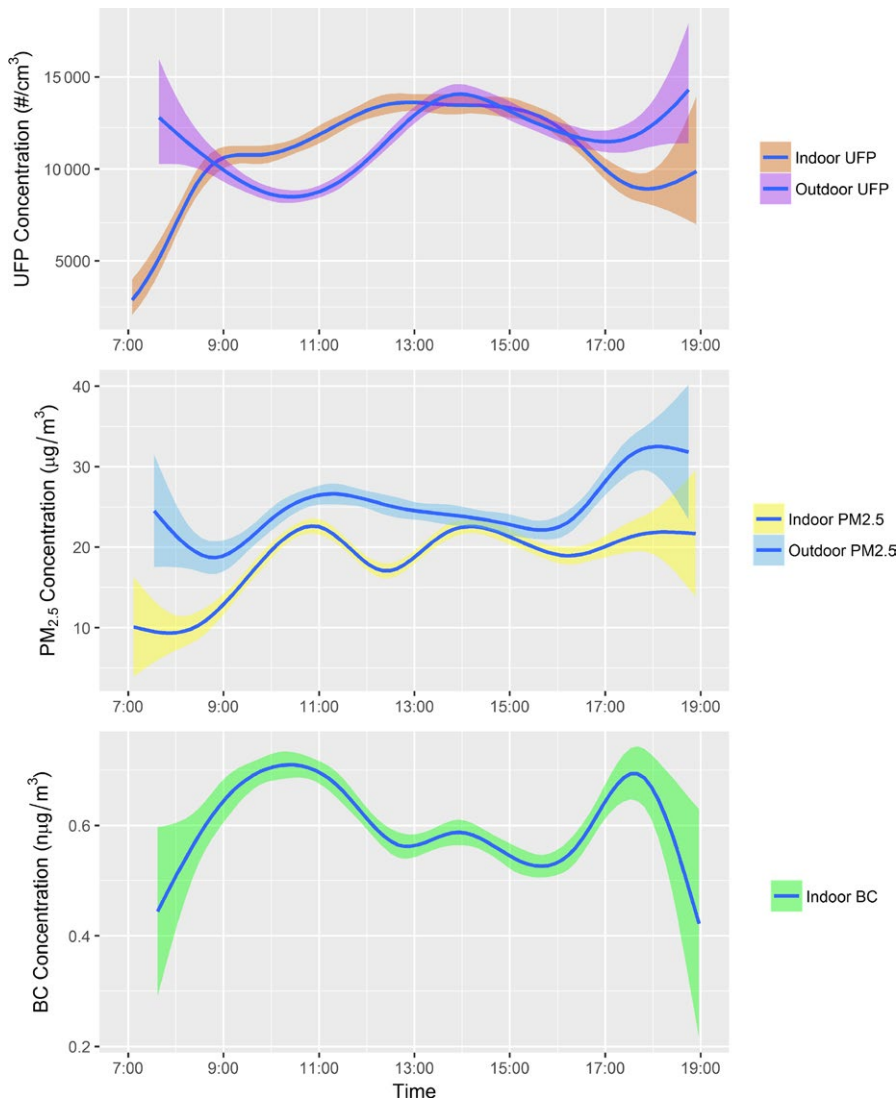


FIGURE 1 Real-time concentrations for indoor and outdoor UFP (top), $PM_{2.5}$ (middle), and black carbon (bottom) through the child-care day. Blue lines and colored bands are expected values and 95% confidence intervals, respectively, per time point from generalized additive models

CAAQS ($PM_{2.5} = 12 \mu\text{g}/\text{m}^3$) and 24-hour NAAQS ($PM_{2.5} = 35 \mu\text{g}/\text{m}^3$) in 66% and 11% of the facilities, respectively. In addition, ECE indoor PM_{10} concentrations exceeded the levels of the annual CAAQS ($PM_{10} = 20 \mu\text{g}/\text{m}^3$) and 24-hour CAAQS ($PM_{10} = 50 \mu\text{g}/\text{m}^3$) in 89% and 46% of the facilities, respectively. No outdoor concentrations exceeded levels of the CAAQS or NAAQS standards. Note, our measurements were collected over ~8 hours, so are not directly comparable to these standards, but indicate that indoor PM exposures in ECE facilities may represent a significant portion of total PM exposure.

4 | DISCUSSION

In this study, we measured multiple metrics of particle pollution indoors and outdoors over the course of a child-care day (~8 hours) at 40 ECE facilities in California. Except for UFP concentrations, most of the particle metrics were highly correlated with each other, indicating common source(s) or determinant(s) of exposure.

Few studies of particulate contamination have been conducted in child-care centers or schools. When compared to these studies, our UFP levels are on the upper range of school exposures; for example, the indoor UFP concentrations (mean = 22 300 particles per cm^3) in our population were higher than mean concentrations measured in six northern California elementary schools (10 800 particles per cm^3 , when children present),⁴⁰ three Ohio schools ($n = 20$ children, 19 800 particles per cm^3),⁴¹ 25 Australian schools (8500 particles per cm^3),⁴² and 39 Spanish schools (15 600 particles per cm^3).⁴³ However, the studies conducted in Australia and Spain used instruments with lower size limits of 10 nm, as compared to the TSI 3781 used in this study with a lower limit of 6 nm, which could potentially underestimate their observed UFP concentrations. Our time series analysis found high mid-day outdoor UFP counts, which may be due to nucleation and cluster growth from photo-oxidized vapors.⁴⁴

Although previous studies have found an association between indoor and outdoor UFP concentrations,^{45,46} we found only a weak, non-significant correlation ($\rho = .35$). This lack of correlation between

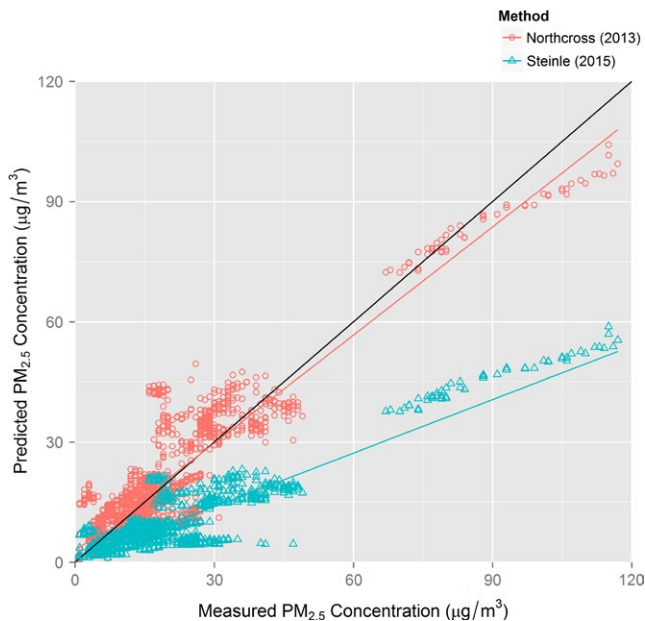


FIGURE 2 Comparison of two methods to convert Dylos $PM_{0.5-2.5}$ count concentrations ($\#/ft^3$) to $PM_{2.5}$ mass concentrations. Black line is the identity line

indoor and outdoor UFP could be related to episodic indoor sources such as cooking appliances. Many of the indoor concentrations were significantly correlated with outdoor concentrations and typically not associated with AER, indicating that outdoor particles are an important source of indoor concentrations, which is consistent with the use of natural ventilation (i.e., open windows) in many of the facilities. Post hoc multiple variable regression confirmed that outdoor concentrations were the best predictors of indoor concentrations and other predictors were typically insignificant. In contrast, the significantly higher indoor PM_{10} concentrations compared to outdoors indicates the presence of indoor sources. None of the variables measured in this study could explain the high levels of indoor PM_{10} , but possible indoor sources might include episodic resuspension during vigorous play or certain arts and crafts activities.

In a small, pilot study conducted in Washington, DC, child-care facilities ($n = 6$), the authors report median real-time $PM_{2.5}$ and PM_{10} concentrations of 18 and 24 $\mu g/m^3$.¹¹ Their $PM_{2.5}$ results are similar to our results, while their PM_{10} concentrations are lower than those found in our study, albeit using different sampling methods (real-time vs gravimetric). Our indoor and outdoor PM_{10} levels were lower than those reported in a study conducted in nine child-care centers in Portugal (total of 102 measurements).⁴⁷ The authors reported median indoor PM_{10} concentrations ranging from 80–126 $\mu g/m^3$ and outdoor PM_{10} concentrations ranging from 60 to 77 $\mu g/m^3$, depending on location in child care and season of sampling. Similar to our results, the authors did not report an association between indoor PM_{10} concentrations and air exchange.

Similar to previous studies,^{26–28} we found the Dylos to be strongly correlated with more expensive real-time particle samplers; therefore, future PM exposure studies could make use of the lower cost Dylos

samplers to assess relative differences in PM concentrations without significant loss of measurement reliability. However, it should be noted that although the Dylos compares well with another optical particle counter, this study and previous studies have shown a significant bias when comparing optical particle counters and gravimetric particle results.^{48,49} Our results only confirm correlation between Dylos and DustTrak measurements.

Our findings suggest that the method used by Northcross et al²⁶ for converting Dylos count concentration to $PM_{2.5}$ mass concentration better predicted the measured $PM_{2.5}$ than the equations presented in Steinle et al²⁸. Of note, the Steinle et al equations did not collect $PM_{2.5}$ using a DustTrak; therefore, the comparisons using a DustTrak in our study may be biased toward the Northcross method that also used a DustTrak to develop their equation.

Indoor gravimetric PM_{10} concentrations exceeded the level of the 24-hour CAAQS in 46% of ECE facilities (16 of 35), and indoor $PM_{2.5}$ concentrations exceeded the level of the 24-hour NAAQS in 11% of ECE facilities (4 of 35; there is no 24-hour CAAQS for $PM_{2.5}$). It should be noted that the measurements in this study were obtained over an 8- to 10-hour period, and do not necessarily represent the levels children were exposed to for a full 24-hour period. However, the monitoring suggests many young children are experiencing a significant portion of their total PM exposures in their child-care facilities and that exposure mitigation may be warranted. There are currently no health-based standards for UFPs or black carbon.

In comparing our indoor black carbon measurements to previously reported levels in the United States, we found our concentrations to be similar to LaRosa et al (2002) but generally lower than those reported.^{21,50,51} In Spanish schools, median indoor black carbon concentrations were 1370 ng/m^3 , approximately three times higher than those found in our study.⁵² Our finding that indoor black carbon concentrations were higher in the urban Alameda County, which includes the Oakland Port and interstate highways with heavy diesel truck traffic,³⁹ and also positively associated with traffic density, supports the hypothesis that nearby traffic is an important source of black carbon exposure to children in ECE facilities. Given that several studies indicate that black carbon is associated with poorer respiratory health in children,^{19–25} and diesel exhaust, the primary source of black carbon in California air,⁵³ is a known carcinogen,⁵⁴ additional research is needed on the contribution of time in child care to total child black carbon exposure.

There are both strengths and limitations to this study. A key strength is the recruitment of a demographically and geographically diverse group of ECE facilities. In addition, we were able to measure multiple particle metrics concurrently both indoors and outdoors. We were also able to quantify a diverse set of potential exposure-level determinants including traffic metrics, ventilation rates, and combustion sources. Although this is the largest study to date examining particles in ECE environments, the relatively small sample size ($n = 40$) limited our statistical power. In addition, measurements collected over a single child-care day may not characterize long-term exposure profiles of these facilities. As our QC results indicate, we occasionally observed large variability between duplicate measurements

including our gravimetric PM measurements and black carbon measurements. For the gravimetric PM, we believe that the variability may have been due to measurements near the detection limits resulting in small absolute differences but large percentage differences. For both the gravimetric PM measurements and black carbon measurements, we still believe the results useful given these metrics were often correlated with other metrics with better QC results. We were unable to control for the potential meteorological and seasonal variability between sampling dates, which could have influenced the particle concentrations observed. Because we compared Dylos and DustTrak measurements at ambient indoor levels, we were not able to test the concentration range in which the correlation observed in this study remained valid. However, Northcross et al found the Dylos had a limit of detection of $<1 \mu\text{g}/\text{m}^3$ and upper limit of $10 \text{mg}/\text{m}^3$ in their controlled experiment.

Finally, our participation rates were low relative to the number of facilities contacted and asked to participate (especially for home-based providers). Participating facilities may have been more interested in environmental exposures than those that chose to not participate, and the results are not necessarily generalizable to other ECE facilities in California.

5 | CONCLUSION

This is the first study to date examining a wide variety of particle metrics in early care and education facilities for young children and has several important implications for future research and public health. Correlations between particle metrics measured in this study indicate that children are receiving co-exposures to the various sizes and composition of particles; therefore, subpopulations of children may be at increased risk of the health effects of early life particulate exposure. Further, as many young children spend a large proportion of their time in ECE facilities, any exposures in these environments are likely to be a significant proportion of their total exposures. Additional research is needed to better understand the contribution of particulate matter exposures in ECE facilities and identify appropriate interventions to mitigate exposures when warranted.

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CONFLICTS OF INTEREST

The authors declare no competing financial interest.

AUTHOR CONTRIBUTIONS

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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SUPPORTING INFORMATION

Additional Supporting Information may be found online in the supporting information tab for this article.

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