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Indoor Exposure to Ambient Particles and Its Estimation Using Fixed Site Monitors

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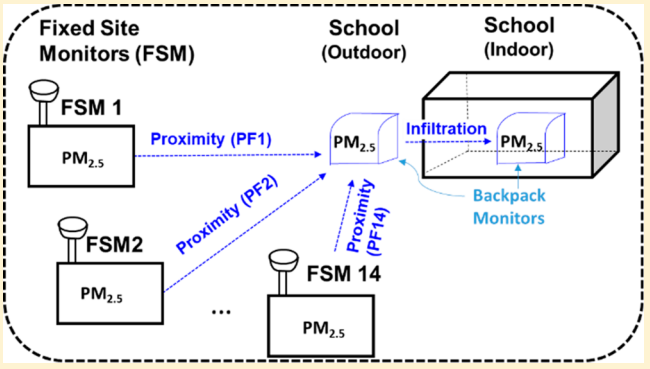
Supporting Information

ABSTRACT: Ambient $PM_{2.5}$ concentrations measured at fixed site monitors (FSM) are often biased with respect to exposure concentrations because of spatial variability and infiltration. Based on comparison of ambient concentrations from 14 FSMs and of exposure concentrations measured indoors and outdoors at two schools in Hong Kong for winter and summer seasons, the magnitude and sources of exposure error based on using FSMs as a surrogate for exposure are quantified. An approach for bias correcting surrogate exposure estimates from FSMs is demonstrated. The approach is based on a proximity factor (PF) that accounts for differences in spatial locations, proximity to emissions and deviation from dominant wind direction, and an infiltration factor (IF) that varies by season. The combination of the PF and IF reduce bias in mean school exposure estimates from ±90% to ±20%. Bias in exposure estimates from using FSMs as surrogates tend to be smaller for which the exposure site and FSM are aligned with wind direction, have similar sampling height, and are in close proximity. The methodology demonstrated to assess concordance between FSMs and exposure measurement sites can be applied more broadly to help reduce exposure error, which may help to interpret seasonal variations in health estimates.



INTRODUCTION

Fine particles, referred to as $PM_{2.5}$, have an aerodynamic diameter of 2.5 μm or less. For ambient $PM_{2.5}$, the weight of evidence is "causal" for cardiovascular effects and premature mortality from both short-term (daily) and long-term (annual) exposure at concentrations typical of air quality in the U.S.[1] Short- and long-term exposures are "likely to be causal" for respiratory effects.[1] The assessment of causality was informed by integration of evidence from multiple health effects studies.[2−9]

Ninety two percent of the world population lives in areas with ambient $PM_{2.5}$ concentrations exceeding the annual World Health Organization (WHO) air quality guideline of 10 $\mu g/m^3$.[10] Human exposure to ambient $PM_{2.5}$ includes exposure to $PM_{2.5}$ concentrations while outdoors, and exposure while indoors or in vehicles to ambient $PM_{2.5}$ that has infiltrated into these microenvironments.[11,12] A microenvironment is a location for which exposure concentrations are homogeneous or well-characterized.[13] $PM_{2.5}$ concentrations recorded at a fixed site monitors (FSM) are typically used as a surrogate for exposure in estimating health effects in epidemiologic studies as they can provide readily available real time data with high measurement accuracy.[14−18] However, measurements from FSMs often lack of spatial coverage and do not account for exposure in different microenvironments.[19,20] Failure to account for the variations in outdoor concentration and outdoor to indoor infiltration may lead to potential for classical exposure error based on differences between exposure and FSM ambient $PM_{2.5}$ concentrations.[13,20,21] Classical exposure

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error leads to effect attenuation and reduced statistical power in epidemiological studies.[19−21]

Most people spend more than 80% of their daily time indoors; therefore, infiltration of ambient $PM_{2.5}$ to indoor microenvironments is an important determinant of personal exposure to $PM_{2.5}$.[22] For example, the infiltration of outdoor $PM_{2.5}$ was found to be up to 80% at homes with open windows.[23] While people are indoors, they are exposed to $PM_{2.5}$ generated from indoor sources and particles infiltrate from outdoors through the building envelope and ventilation systems. In enclosed microenvironments, the contribution of ambient air pollution to indoor exposure can be quantified using linear regressions between simultaneous indoor and outdoor measurements.[11,24−26] The infiltration factor (IF), which is the equilibrium fraction of ambient particles that infiltrate indoors and remain suspended, is inferred as the slope from linear regression.

Infiltration factors derived from linear regression are widely used in microenvironmental (ME) models, such as the U.S. Environmental Protection Agency (EPA) Air Pollutants Exposure (APEX) model or Stochastic Human Exposure and Dose Simulation (SHEDS-PM) model for PM.[12,27] In these ME models, the IFs are used to estimate the exposure to ambient pollutants in indoor microenvironments based on outdoor ambient concentrations.

$PM_{2.5}$ concentrations from FSMs are commonly used in these models as a surrogate for outdoor concentration.[12,28,29] However, the use of FSM outdoor concentrations may under- or overestimate the outdoor concentrations nearby the targeted microenvironment due to spatial variability in outdoor $PM_{2.5}$ concentrations.[30−33] For example, Raysoni et al. found that the outdoor $PM_{2.5}$ concentrations measured at four schools were 15−40% higher than at the nearest FSMs.[33] In some epidemiological studies, exposure differences due to spatial variations in outdoor concentration have been considered categorically based on their proximity to a major emission source. For example, a cross-sectional study conducted in Italy categorized the subjects into highly exposed, moderately exposed, and unexposed groups based on their distance from a major road.[34] In APEX, a proximity factor (PF) is used to account for spatial differences in outdoor $PM_{2.5}$ concentrations between a FSM and the outdoors adjacent to the microenvironment of interest. PFs can be derived from linear regression of $PM_{2.5}$ concentrations between outdoors adjacent to the targeted microenvironments and the selected FSMs.[27] However, studies on PF are scarce. If data on PF are unavailable, a default PF value of one is typically used, which may lead to classical errors in exposure estimates.

Recent studies indicate that spatial differences in outdoor $PM_{2.5}$ concentrations are related to geographical and meteorological parameters.[35−38] For example, Eeftens et al. found that 21−79% of the variability in annual average outdoor $PM_{2.5}$ concentration in 20 European study areas can be explained by differences in GIS-derived predictor variables, such as elevation, X/Y-coordinate and land use information.[35] These factors reflect geographic location characteristics which may relate to the dispersion of pollution, distance to the emission sources, and intensity of nearby emissions.[35] Wind direction is also an important variable that determines the impact of sources on receptor points.[26,39−41] For example, the outdoor $PM_{2.5}$ concentration at 10 schools in Pakistan were observed to be 20−35% higher in winter than in summer due to differences in dominant wind direction.[42] Quantifying the association between influential sources of spatial variability and the PF may help in evaluating whether a FSM is a useful outdoor surrogate for a targeted location and help in developing methods to address the classical error in exposure estimates for indoor microenvironments.

There are 16 FSMs spatially distributed over an area of 1100 km^2 in Hong Kong, of which 14 stations have long-term available monitoring data. The accessibility of air quality monitoring data from these stations provides a unique opportunity to investigate the errors in exposure estimates that accrue from using FSM measured concentrations as a surrogate for outdoor concentrations adjacent to a microenvironment of interest. School is an important microenvironment because children of school age are particularly susceptible to exposure to $PM_{2.5}$, and exposure at schools may impose health risks and adversely affect study performance.[43−47] However, studies on children's exposure to ambient particles at schools are scarce.[28] Two schools, located in areas with different location characteristics, were chosen for investigation. Indoor and outdoor measurements were made at both schools in both winter and summer, which enables quantification of infiltration factors and proximity factors under different conditions. The objectives of this paper are to (1) assess the sensitivity of PF to the choice of FSM; (2) quantify the exposure error introduced by using FSM as an outdoor surrogate with and without PF correction; and (3) determine whether variability in the PF can be explained by location characteristics.

MATERIALS AND METHODS

The methodology includes identification of factors affecting outdoor $PM_{2.5}$ concentrations and infiltration of $PM_{2.5}$ into indoors, site selection, measurement of indoor and outdoor exposure concentrations at selected schools, analysis of FSM data, quantification of IF and PF derived from linear regression models, development of exposure error under three scenarios, development of concordance indices and quantification of their associations with PF, and statistical methods in data analysis.

Site Selection. Factors affecting spatial variations in outdoor $PM_{2.5}$ concentrations and $PM_{2.5}$ infiltration were considered in deciding when and where to measure indoor and outdoor $PM_{2.5}$ concentrations.

Outdoor $PM_{2.5}$ concentrations vary with space and time due to variations of emission sources and meteorology.[35,36,38,39] Concentrations measured at a specific location are related to its proximity to local sources, geographic location relative to the air pollution flow determined by the wind, and terrain features regarding air pollution dispersion.[35,36,39,48] As an example of the influence of proximity to emission sources, the annual $PM_{2.5}$ concentrations monitored at a roadside site in Hong Kong were nearly 2.5 times higher than at a rural background site.[49] With regard to direction of air flow, outdoor air quality in Hong Kong is closely related to the Asian Monsoon system, characterized by shifts in dominant wind direction. The persistent northeast monsoon in winter brings pollutants from the Asian continent, whereas the summer monsoon shifts to southwesterly winds that bring in cleaner marine air.[49−51] The average $PM_{2.5}$ concentrations observed at a background site in Hong Kong were 2 times higher in winter than in summer.[50] Air dispersion tends to be better at higher elevation, whereas exposure microenvironments located in a valley or urban street canyon may experience higher ambient concentrations.[38,48]

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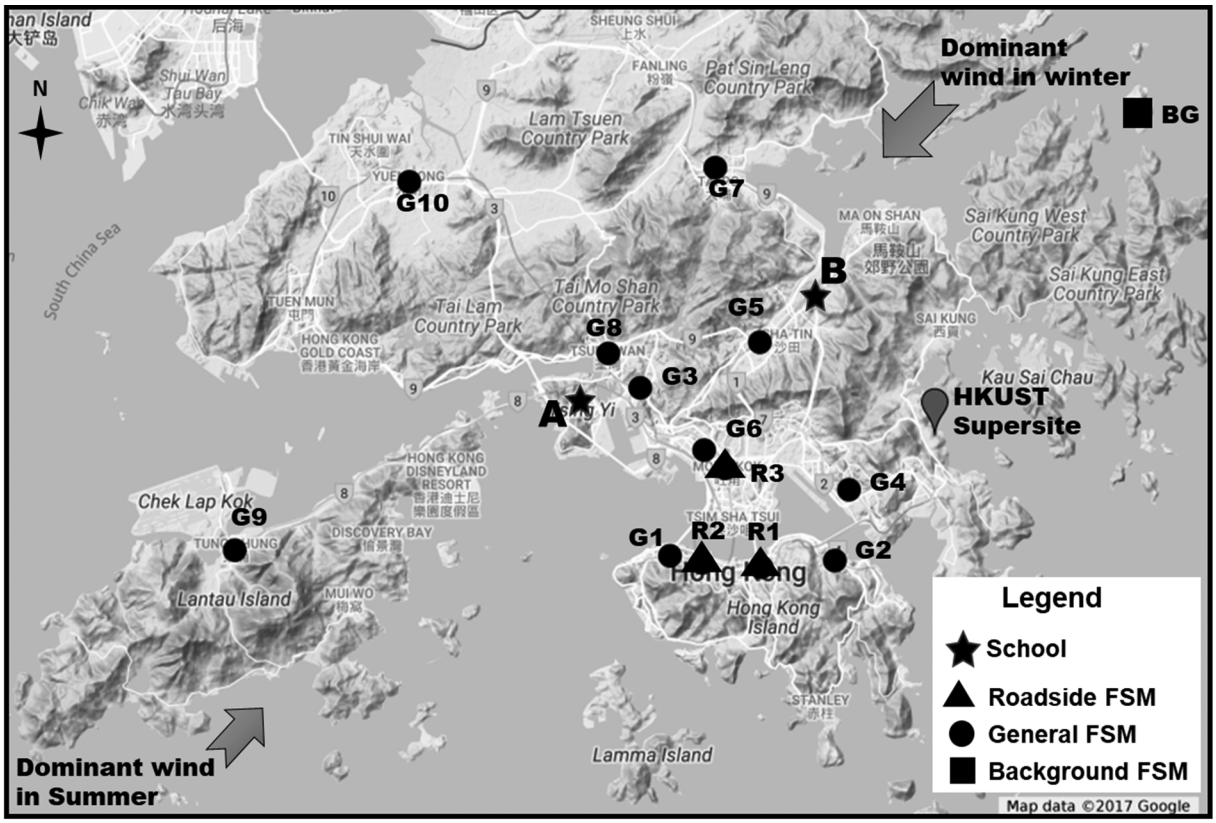

Note:

[a.] The background topographic map is obtained from Google map.

[b.] Distance from FSM sites to selected schools (km)

FSM Site	R1	R2	R3	G1	G2	G3	G4	G5	G6	G7	G8	G9	G10	BG
to school A	12.2	10.2	8.1	9.1	14.8	3.2	13.9	9.2	6.9	12.7	2.5	17.5	12.7	29.9
to school B	12.5	13.1	8.6	13.4	12	8.9	8.8	2.9	8.4	8.1	9.8	29.5	20	18.1

Figure 1. Geographic location of indoor and outdoor at selected schools and the 14 fixed-site monitors (FSM) in Hong Kong.

The selected 14 FSMs are spread widely over the Hong Kong territory, including three roadside sites (R1-R3), one background site (BG), and ten general stations (G1-G10). The roadside and general stations are located in urban areas. The background site is located in a northeast rural area, which is upwind of other sites in winter. Most of the FSMs have an elevation lower than 15 m, except the site BG (16 m) and G1 (67 m). The height of the sampling inlet from ground at these sites ranges from 3 to 4.5 m at the roadside monitors, to 11 to 27.5 m at the background and general sites. Roadside monitors are more subject to local vehicle emissions because of their close proximity to the road (less than 5 m) and lower sample inlet height.[49]

Infiltration of outdoor particles depends on ventilation.[23,52−54] For example, IF was found to be approximately 0.1 at homes with air conditioning (A/C) on and 0.8 at homes with open windows.[23] Common ventilation methods at schools in Hong Kong include natural ventilation through open windows and doors, and mechanical ventilation through the use of inlet and exhaust fans, ceiling fans and A/C.[55] Typically, natural ventilation is used in winter. A/C is used in summer when ambient temperature is higher than 25 °C.[55]

Two public primary schools were selected: one on a hillside with an elevation of 82 m (School A) and the other in a valley with an elevation of 6 m (School B). They are shown in Figure 1. Public schools in Hong Kong are operated or aided by the government and provide free education for more than 80% of the total primary students.[56,57] The selected schools are typical public schools in Hong Kong with 6-stories of mostly classrooms and an average class size of 30 students.[57] There are no obvious combustion sources inside the selected schools as meals are provided by lunchboxes delivered from suppliers outside campus, which is the common practice in Hong Kong.[58] School A is adjacent to a road with annual average traffic flow of less than 500 vehicles/hour.[59] School B is located in a congested urban area adjacent to a road with annual average traffic flow of 1500 vehicles/hour.[59] Both schools use natural ventilation in winter with open windows and doors, and A/C with ceiling fans in summer. Windows and doors were usually closed during A/C use, except that some windows at School B were $^{1}/_{10}$−$^{1}/_{8}$ of fully open in summer to help reduce a flu epidemic.

School Measurement and Quality Control. Measurements at the selected schools were conducted during the

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school semester under normal operation with students in attendance. Continuous indoor and outdoor measurements were simultaneously conducted on weekdays during school hours (8:00 a.m. to 3:00 p.m.). $PM_{2.5}$ concentrations measured in both classrooms and nonclass rooms, such as assembly hall and multifunction room, were used to quantify indoor concentrations, which reflect the exposure concentrations during typical school activities. Outdoor $PM_{2.5}$ concentrations were measured along a transect along the nearest road, which was less than 0.5 km from the school. The sampling periods ranged from January 5 to 30, 2015 in winter and from June 1 to 28, 2015 in summer.

$PM_{2.5}$ concentrations were measured using a portable light scattering laser photometer, TSI DustTrak II Aerosol Monitor 8530.[60,61] CO_2 concentrations and temperature and relative humidity were measured using a TSI Q-Trak model 7575.[62] All measurements were made at 1 Hz. CO_2 is an indicator of ventilation and human occupancy in enclosed spaces.[63] Two identical sets of instruments were placed into two backpacks, each with one DustTrak and one Q-Trak, for simultaneous measurements indoors and outdoors at each school. The sampling tube outside the backpack was placed at the height of nose level of a student while sitting in the classroom, which was approximately 0.9 m above the ground.

Quality assurance included instrument calibration, time synchronization, a before-and-after check list, consistency checks, cross-checking during parallel measurements, and measurement protocol. The DustTrak measured $PM_{2.5}$ concentrations are typically a factor of 2−4 higher compared to reference methods, such as those used in FSMs.[61,64,65] The light scattering detection method used in the DustTrak is potentially sensitive to humidity, which affects particle size and shape.[66,67] To enable comparison with FSMs, the DustTraks were calibrated against a federal equivalent method, a Thermo Fisher Scientific Synchronized Hybrid Ambient Real-time Particulate monitor, SHARP model 5030,[68] at the Hong Kong University of Science and Technology (HKUST) air quality research supersite. Calibration factors were derived for each DustTrak based on 7−8 h of colocation with the SHARP at 1 min resolution in each winter and summer period, to account for seasonal differences in humidity.

All instruments were time synchronized each day to the Hong Kong Observatory clock. The DustTrak and Q-Trak were zero calibrated in the lab before each measurement. A before-and-after checklist was developed to record instrument operation status. The two backpacks were cross-checked in parallel by colocating them during daily trips between the lab at HKUST and the schools. A detailed measurement standard operating procedure with log sheets was developed.

Fixed Site Monitors. Hourly average ambient $PM_{2.5}$ concentrations were obtained from each FSM during the sampling period in each season. The $PM_{2.5}$ concentrations at the selected FSMs were measured with hourly or subhourly resolution using "reference methods" or "equivalent methods" designated by the U.S. EPA, including Thermo Scientific Partisol-Plus 2025, R&P TEOM Series 1400a-AB-$PM_{2.5}$, Thermo Scientific TEOM 1405-DF, Met One BAM1020, and T-API 602 Beta Plus.[69] Hourly wind speed and wind direction were obtained from a background meteorological station (Waglan Island) in Hong Kong. The average wind speed was 6.1 m/s in winter with stand deviation of 2.6 m/s, and 5.5 m/s in summer with stand deviation of 2.1 m/s. The dominant wind direction in Hong Kong is northeast in winter and south to southwest in summer.

Factors that may affect the differences in outdoor $PM_{2.5}$ concentrations between schools and FSMs were hypothesized to include distance between the school and FSM, differences in elevation and sampling height between the school and FSM, and direction from the school to the FSM relative to the dominant wind direction. These comparative variables were quantified based on site data for each school and FSM.

Infiltration Factor and Proximity Factor. Infiltration factors for outdoor particles at selected schools were derived based on linear regression between simultaneous indoor measurements at a school and outdoor measurements made in close proximity to the school, which was at the nearby road transect, in each season:

$$C_{\mathrm{idr},s,p,t} = \mathrm{IF}_{s,p} C_{\mathrm{odr},s,p,t} + C_{\mathrm{NA},s,p,t} + \varepsilon_t \quad (1)$$

Where $C_{\mathrm{idr},s,p,t}$ = indoor $PM_{2.5}$ concentrations at school s in season p at time t ($\mu g/m^3$); $C_{\mathrm{odr},s,p,t}$ = outdoor $PM_{2.5}$ concentrations at school s in season p at time t ($\mu g/m^3$); $C_{\mathrm{NA},s,p,t}$ = nonambient component of indoor concentration at school s in season p at time t ($\mu g/m^3$); $\mathrm{IF}_{s,p}$ = infiltration factor derived for school s in season p, unitless; ε_t = random error ($\mu g/m^3$); idr = indoor location (e.g., classroom); s = school index (e.g., School A, School B); p = season (e.g., winter, summer); t = time step (hour); odr = outdoor location in close proximity to the school, which is the nearest transect in this study; NA = nonambient sources.

PFs were estimated by school and season based on linear regression models (LRM) developed from outdoor $PM_{2.5}$ concentrations between the nearest transect and each of the FSMs:

$$C_{\mathrm{odr},s,p,t} = \mathrm{PF}_{s,\mathrm{FSM},p} C_{\mathrm{odr,FSM},p,t} + C_{\mathrm{UE},s,\mathrm{FSM},p,t} + \varepsilon_t \quad (2)$$

Where $C_{\mathrm{odr,FSM},p,t}$ = outdoor $PM_{2.5}$ concentrations measured at a FSM in season p at time t ($\mu g/m^3$); $C_{\mathrm{UE},s,\mathrm{FSM},p,t}$ = unexplained concentrations at school s that is not explained by the variability in the concentration at a FSM in season p at time t ($\mu g/m^3$); $\mathrm{PF}_{s,\mathrm{FSM},p}$ = Proximity factor estimated between school s and a FSM in season p, unitless; FSM = fixed site monitor, i.e., R1-R3, G1-G10, BG; UE = unexplained sources which may associated with local sources.

Exposure Error. To assess the exposure error introduced from using different outdoor surrogates, indoor $PM_{2.5}$ concentrations were estimated based on three sources of direct or surrogate outdoors concentrations: (S1) outdoor concentration measured at transects in close proximity to the selected school; (S2) outdoor concentration from a FSM; and (S3) outdoor concentration from an FSM adjusted by an appropriately calibrated $\mathrm{PF}_{s,\mathrm{FSM},p}$. In all three scenarios, $\mathrm{IF}_{s,p}$ is used to estimate indoor concentration based on outdoor concentration.

The exposure errors introduced by using different outdoor surrogates were evaluated by comparing the differences between hourly estimated indoor $PM_{2.5}$ concentrations and the hourly measured indoor concentrations for both schools in both winter and summer:

$$\mathrm{EE}_{r,s,p,t} = \frac{(C_{r,s,p,t} - C_{\mathrm{idr},s,p,t})}{C_{\mathrm{idr},s,p,t}} \times 100\% \quad (3)$$

Where $\mathrm{EE}_{r,s,p,t}$ = exposure error under scenario r at school s in season p at time t, unitless; $C_{r,s,p,t}$ = estimated concentration

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Table 1. Spearman Correlation Coefficient between Concordance Indices and PF, and Final Model[a]

concordance indices (CI)			with PF		
name	description	unit	all (N = 56)	School A (N = 28)	School B (N = 28)
$D_{s,\ FSM}$	distance between school and FSM	km	0.01	0.00	0.00
$IVSD_{s,FSM}$	inverse $D_{s,FSM}$		−0.01	0.00	0.00
$logD_{s,FSM}$	log of $D_{s,FSM}$		0.01	0.00	0.00
$\Delta E_{s,FSM}$	the differences in elevation between school and the FSM	m	0.12	0.10	0.09
$IVS\Delta E_{s,FSM}$	inverse $\Delta E_{s,FSM}$		0.01	−0.10	−0.06
$log\Delta E_{s,FSM}$	log of $\Delta E_{s,FSM}$		−0.08	−0.10	0.11
$\Delta H_{s,FSM}$	difference in elevation and sampling height between school and FSM	m	0.05	0.06	0.04
$IVS\Delta H_{s,FSM}$	inverse $\Delta H_{s,FSM}$		−0.05	−0.06	−0.04
$log\Delta H_{s,FSM}$	log of $\Delta H_{s,FSM}$		0.05	0.06	0.04
$\Delta W_{s,FSM}$	the angle difference between school to FSM and dominant wind direction	radian	**0.67**	**0.59**	**0.71**
$IVS\Delta W_{s,FSM}$	inverse $\Delta W_{s,FSM}$		**0.33**	**0.42**	0.27
$log\Delta W_{s,FSM}$	log of $\Delta W_{s,FSM}$		**0.33**	−0.49	**0.67**
MIX1	$logD_{s,\ FSM} \times \Delta W_{s,FSM}$		**0.64**	**0.53**	**0.70**
MIX2	$log\Delta E_{s,FSM} \times \Delta W_{s,FSM}$		**0.49**	**0.57**	0.46
MIX3	$log\Delta H_{s,FSM} \times \Delta W_{s,FSM}$		**0.62**	**0.57**	**0.66**
MIX4	$logD_{s,\ FSM} \times log\Delta E_{s,FSM} \times \Delta W_{s,FSM}$		**0.47**	**0.51**	0.46
MIX5	$logD_{s,\ FSM} \times log\Delta H_{s,FSM} \times \Delta W_{s,FSM}$		**0.60**	**0.53**	**0.68**
MIX6	$log\Delta E_{s,FSM} \times log\Delta H_{s,FSM} \times \Delta W_{s,FSM}$		**0.50**	**0.56**	0.46
MIX7	$logD_{s,FSM} \times LOG\Delta E_{s,FSM} \times log\Delta H_{s,FSM} \times \Delta W_{s,FSM}$		**0.48**	**0.52**	0.46
final model:	$PF_{s,FSM} = (0.29 \pm 0.06) \times \Delta W_{s,FSM} - (0.02 \pm 0.01) \times MIX5 + (0.54 \pm 0.06)$ (N = 56; R^2 = 0.53; AIC = −43.2)				

[a]Note: bold figures are estimates with p-value < 0.05.

under scenario r at school s in season p at time t ($\mu g/m^3$); r = scenarios, i.e., S1, S2, S3.

Concordance Indices. Four concordance indices (CIs) were developed based on factors that may affect the spatial variability of $PM_{2.5}$ concentrations, including distance between school and FSM ($D_{s,FSM}$), difference in elevation ($\Delta E_{s,FSM}$) and sampling height ($\Delta H_{s,FSM}$) between school and FSM, and the angle difference between school to FSM and the dominant wind direction ($\Delta W_{s,FSM}$). $D_{s,FSM}$, and $\Delta E_{s,FSM}$ represent the horizontal and vertical spatial differences between the selected schools and FSMs. $\Delta H_{s,FSM}$ represents the vertical proximity to ground emission sources, whereas $\Delta W_{s,FSM}$ represents the geographic location relative to the air pollution flow. Different forms of these indexes, such as inverse and log, are compared to examine possible relationships under different weightings on CIs.[35] The inverse forms are developed based on assumed Gaussian dispersion to reflect the inversely decreasing impact from emissions with distance and height.[70,71] The log-normal forms take into account that the variability in the quantified variable must be non-negative. To investigate the interactions among these factors and their impacts on PF, seven mixed CIs were introduced based on various combinations of products of individual CIs, as listed in Table 1. To assess the sensitivity of associations to school location, the associations between $PF_{s,FSM,p}$ and each CI were examined separately for School A and School B. Associations between $PF_{s,FSM,p}$ and the CIs were used to develop guidance regarding whether a FSM is a useful surrogate for ambient air quality immediately outside the selected school. The relationship between $PF_{s,FSM}$ and CIs were quantitatively described using a linear regression model to enable inference of the magnitude of the PF for sites of interest.

Statistical Analysis. Data from the portable monitors were merged into a 1 min averaging time data set using SAS version 9.3. The 1 min average $PM_{2.5}$ data were scaled based on the calibration factors derived for each DustTrak in comparison to the SHARP at the same time resolution in each season. For comparison to FSMs, the 1 min DustTrak $PM_{2.5}$ concentrations were averaged on an hourly basis. Association between indoor and outdoor $PM_{2.5}$ concentrations at schools and FSMs were quantified using Spearman correlation coefficient (SCC), which assesses how well the relationship between two variables can be described using a monotonic function.[72] The association assessed by SCC can be either linear or nonlinear.

Estimates and standard errors of $IF_{s,p}$ and $PF_{s,FSM,p}$ were derived from LRMs as given in eqs 1 and 2, respectively. Two tailed t tests were used to determine whether the slopes and intercepts estimated from LRMs were significantly different from zero and whether the slopes were significantly different from unity at a 95% confidence level (p-value < 0.05). The developed LRMs were evaluated based on the coefficient of determination (R^2), which indicates the proportion of the variance in the dependent variable that can be explained by the independent variable. Associations between $PF_{s,FSM,p}$ and individual CIs were investigated using SCC. The quantitative relationship between $PF_{s,FSM,p}$ and CIs with significant SCC was investigated using a stepwise regression model with entry and stay significance level of 0.1. The final model for $PF_{s,FSM,p}$ was determined by the minimized Akaike's information criteria (AIC). Differences in indoor $PM_{2.5}$ concentrations were calculated between hourly estimated indoor concentrations and the corresponding measured values for each school and season. For each outdoor concentration scenario (S1, S2, S3), exposure errors were quantified based on their mean and standard deviation, and were visualized with boxplots.

Continuous measurements have been reported to be autocorrelated, indicating serial dependence with previous observations.[73] The presence of autocorrelation is a potential factor that may affect the regression analysis between outcome and explanory variables.[74] Sensitivity tests were conducted to evaluate the impact of autocorrelation on the infiltration estimates. For a 1 h time interval, there is no significant differences in IF estimates between with and without

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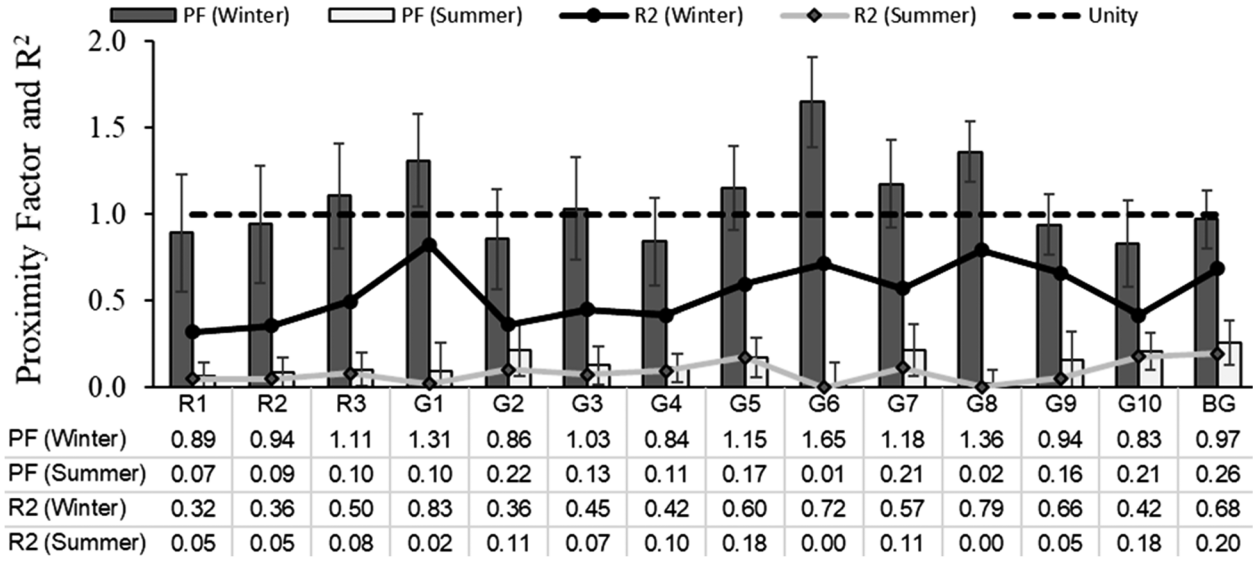
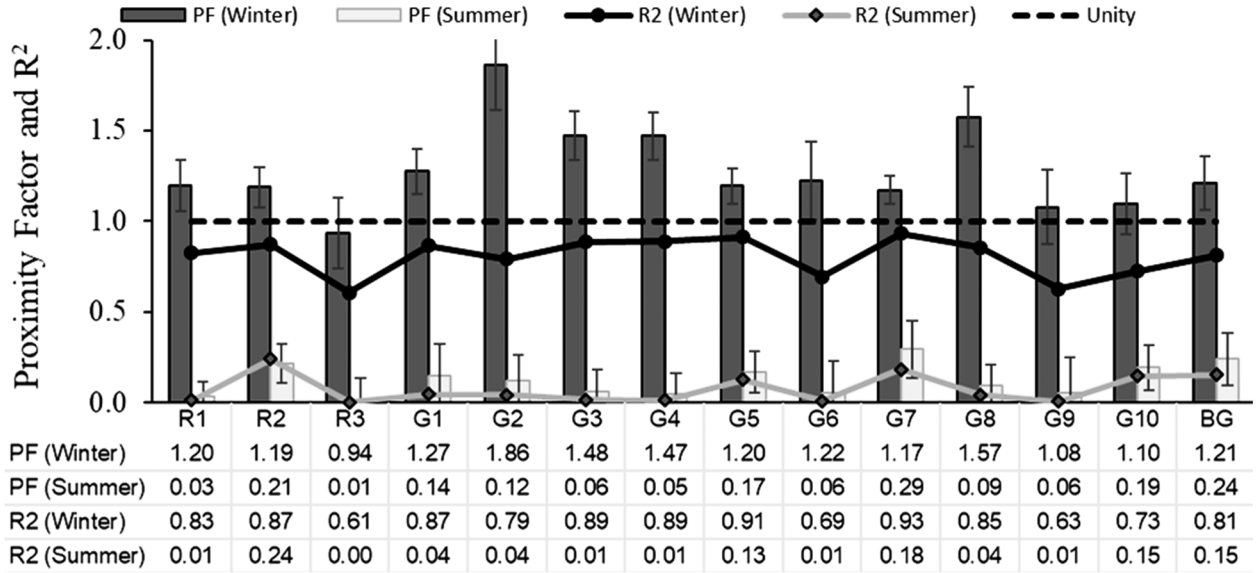


	R1	R2	R3	G1	G2	G3	G4	G5	G6	G7	G8	G9	G10	BG
PF (Winter)	0.89	0.94	1.11	1.31	0.86	1.03	0.84	1.15	1.65	1.18	1.36	0.94	0.83	0.97
PF (Summer)	0.07	0.09	0.10	0.10	0.22	0.13	0.11	0.17	0.01	0.21	0.02	0.16	0.21	0.26
R2 (Winter)	0.32	0.36	0.50	0.83	0.36	0.45	0.42	0.60	0.72	0.57	0.79	0.66	0.42	0.68
R2 (Summer)	0.05	0.05	0.08	0.02	0.11	0.07	0.10	0.18	0.00	0.11	0.00	0.05	0.18	0.20

(a) School A



	R1	R2	R3	G1	G2	G3	G4	G5	G6	G7	G8	G9	G10	BG
PF (Winter)	1.20	1.19	0.94	1.27	1.86	1.48	1.47	1.20	1.22	1.17	1.57	1.08	1.10	1.21
PF (Summer)	0.03	0.21	0.01	0.14	0.12	0.06	0.05	0.17	0.06	0.29	0.09	0.06	0.19	0.24
R2 (Winter)	0.83	0.87	0.61	0.87	0.79	0.89	0.89	0.91	0.69	0.93	0.85	0.63	0.73	0.81
R2 (Summer)	0.01	0.24	0.00	0.04	0.04	0.01	0.01	0.13	0.01	0.18	0.04	0.01	0.15	0.15

(b) School B

Note:

[a] See Table S1 for additional information on each monitor;

[b] PF refers to proximity factor derived based on linear regression of $PM_{2.5}$ concentrations between transects in close proximity to the selected school and the FSMs;

[c] The figures for PF given in the table is the mean PF value;

[d] R^2 refers to coefficient of determination of the linear regression models for the derived PF.

Figure 2. Proximity factor and its confidence interval derived for fixed site monitors at school A and B.

autocorrelation correction. Details are provided in the Supporting Information.

RESULTS

Sixty to seventy sampling hours were completed for each school and each season. Hourly data were obtained from 14 FSMs during the school sampling periods. The results include instrument calibration, measurement data summary, estimation of infiltration factors, estimation of proximity factors, quantification of exposure error for three outdoor surrogate concentration scenarios, and association betweenproximity factors and proposed concordance indices.

Calibration. The calibration factors for DustTraks 1 and 2 were 2.72 (R^2 = 0.98) and 2.67 (R^2 = 0.98) in winter and 3.26 (R^2 = 0.87) and 3.02 (R^2 = 0.91) in summer, respectively. The average relative humidity (RH) and temperature during the calibration were 49% and 17 °C in winter and 53% and 29 °C in summer, respectively. The absolute humidity was 9 g/m^3 in winter and 20 g/m^3 in summer, respectively. The higher humidity in summer favors the growth of particles which leads

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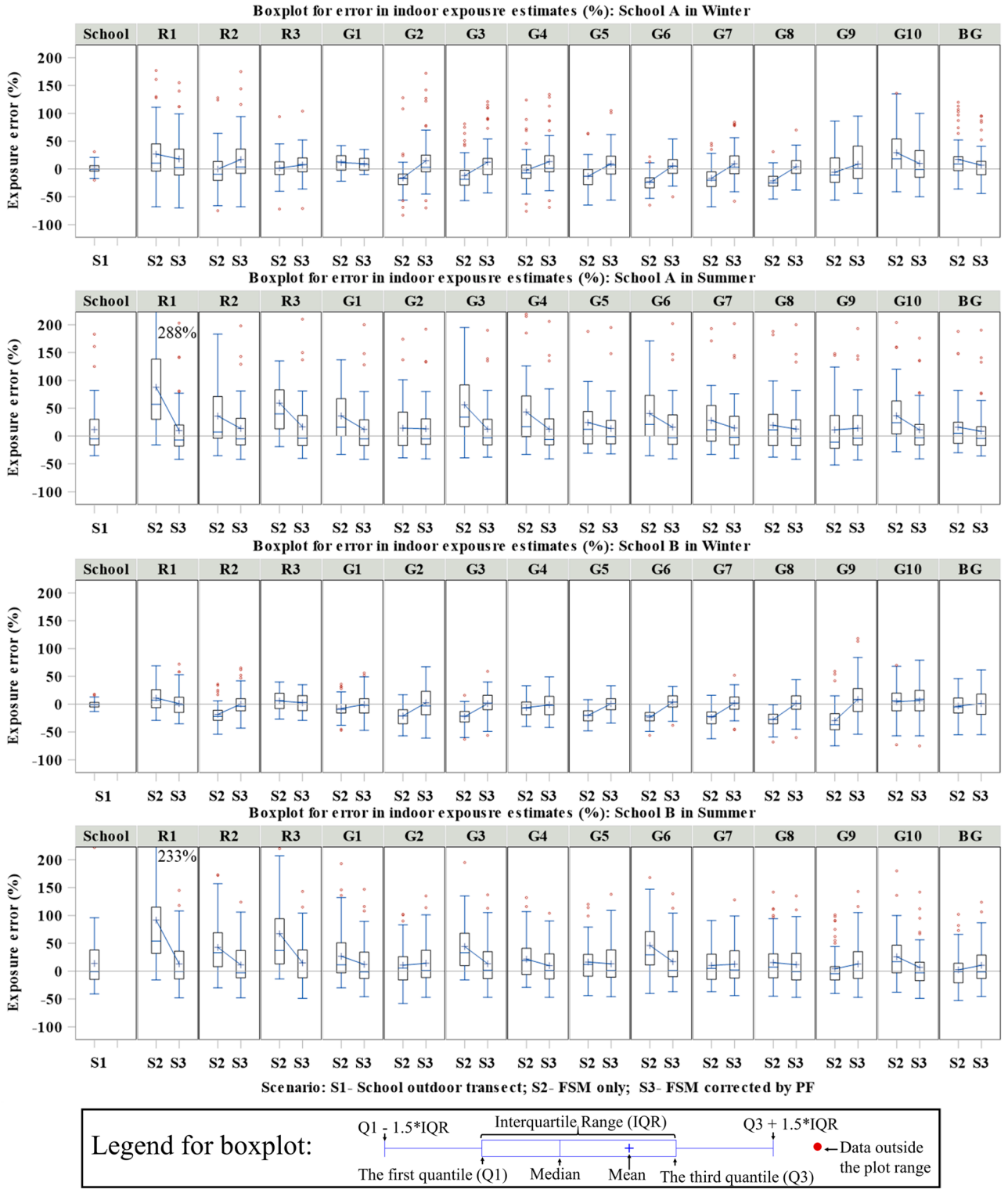

Figure 3. Boxplot of the estimate error in hourly indoor exposure concentration by school and season using (S1) outdoor concentration measured at transects, (S2) FSM concentration, and (S3) PF corrected FSM concentration.

to a higher calibration factor.[66,67] These season-specific calibration factors are used for the subsequent data analysis. The accuracy of calibration of the gas sensors was confirmed based on measurements with standard gas at a known concentration in the lab.

Summary of Measurements. The winter average outdoor $PM_{2.5}$ concentrations were 43 $\mu g/m^3$ and 63 $\mu g/m^3$ at Schools A and B, respectively. The average $PM_{2.5}$ concentrations recorded at the FSMs ranged from 25 $\mu g/m^3$ at G1 to 67 $\mu g/m^3$ at R1. Significantly high correlations in $PM_{2.5}$ concentrations were found among all outdoor locations, including between FSMs and school transects, with most of the SCC values being higher than 0.7. The prevailing winds in winter were northeasterly. The high SCCs values indicate the

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influence of a regional pollution plume on the FSMs and schools.

The summer average outdoor $PM_{2.5}$ concentrations were 7 $\mu g/m^3$ at both schools. The average $PM_{2.5}$ concentrations recorded at the FSMs ranged from 6 $\mu g/m^3$ at G9 to 22 $\mu g/m^3$ at R1. The correlations in outdoor $PM_{2.5}$ concentrations between each pairing of an FSM and transect at a school were generally weak, with most of the SCC values being insignificant and lower than 0.3. This indicates that the summer outdoor $PM_{2.5}$ concentrations in Hong Kong are most affected by local sources. Site R1 recorded the highest $PM_{2.5}$ concentrations among all FSMs in both winter and summer, due to its proximity to traffic emission at curbside with the lowest sampling height of 3 m.

Infiltration Factors. The winter indoor $PM_{2.5}$ concentrations were highly correlated with outdoor $PM_{2.5}$ concentrations measured at transects at both schools (SCC > 0.98), with average differences between indoor and outdoor concentrations of less than 5%. Natural ventilation with open windows was used at both schools in winter. The mean and standard error of estimated $IF_{s,p}$ in winter were 0.91 ± 0.02 at School A and 0.97 ± 0.02 at School B, respectively. The outdoor $PM_{2.5}$ concentrations at transect explained 98% (R^2 = 0.98) and 99% (R^2 = 0.99) of the variability in indoor concentration at Schools A and B, respectively. The winter $IF_{s,p}$ at School B was not significantly different than unity, indicating there is no difference in exposure to ambient $PM_{2.5}$ whether indoors or outdoors at that school while windows were open. A regional air pollution episode of unusually high $PM_{2.5}$ concentrations was observed on January 21th during the measurement at School B. During this episode, the mean $PM_{2.5}$ concentration was 138 $\mu g/m^3$ at the background FSM (BG) and 152 $\mu g/m^3$ at the outdoor transect. The mean indoor $PM_{2.5}$ concentration measured on that day was 148 $\mu g/m^3$, which was substantially higher than the average indoor concentrations of 49 $\mu g/m^3$, for all other measured days.

The summer indoor $PM_{2.5}$ concentrations measured at schools A and B were weakly correlated with those measured at outdoor transects, with SCC values less than 0.35 at both schools. The summer $IF_{s,p}$ was 0.27 ± 0.13 at School A and 0.25 ± 0.09 at School B. A/C was used in both schools in summer.

The average indoor CO_2 concentrations were 430 and 400 ppm at Schools A and B, respectively, in winter, and 920 and 720 ppm, respectively, in summer. The number of students at each school did not vary between seasons, indicating that exhalation of CO_2 was similar between winter and summer. The substantially higher indoor CO_2 concentrations observed in summer indicate reduction in air exchange associated with the use of A/C. The reduction in air exchange helped to decrease the infiltration of outdoor particles. The outdoor $PM_{2.5}$ concentrations at the transects explained only 7% (R^2 = 0.07) and 13% (R^2 = 0.13) of the variability in summer indoor concentration at Schools A and B, respectively.

Proximity Factors. Estimates of $PF_{s,FSM,p}$ for both schools, both seasons, and all 14 FSMs are presented in Figure 2, together with coefficients of determination (R^2). In winter, variability in FSM concentrations were medium to strong predictors of variability in transect concentrations for School A. The R^2 ranged from 0.32 to 0.83 among FSMs. The FSMs with the highest R^2 were G1 (R^2 = 0.83), a site with elevation and sampling height comparable to School A, and G8 (R^2 =0 .79), the nearest upwind site only 2.5 km away. The lowest R^2 of 0.32 was found at R1, a roadside monitor located off to the side of the dominant wind corridor that affects School A.

The $PF_{A,FSM,w}$ estimates for School A in winter ranged from 0.83 to 1.65 among FSMs. Most of the $PF_{A,FSM,w}$ values were not significantly different than unity except for PFs of 1.36 at G1, 1.65 at G6, and 1.31 at G8. The intercepts of LMRs were not significantly different from zero at these three sites, therefore, $PF_{A,FSM,w}$ values higher than unity indicate that transect concentrations would be underestimated at School A if $PM_{2.5}$ concentrations at these sites were used as surrogates. For example, the winter average $PM_{2.5}$ concentration at G8 was 32% lower than at the School A transect which is close to the bias in $PF_{A,G8,w}$ for this FSM, $[PF_{A,G8,w} - 1]$, of 0.31. The intercept was significantly higher than zero at G2, a site located off to the side of the dominant wind corridor that affects School A, indicting a contribution from local sources.

Variability in winter $PM_{2.5}$ concentrations at FSMs explains 61% to 93% of the variability in outdoor transect concentrations at School B. The highest R^2 was found at G7 (R^2 = 0.93) and G5 (R^2 = 0.91), the nearest FSMs, whereas the lowest R^2 was found at G9 (R^2 = 0.63), the most distant FSM, and R1 (R^2 = 0.61), a roadside FSM. Most of the FSMs were located downwind of School B in winter. For 5 out of 14 FSMs, the $PF_{B,FSM,w}$ values were significantly higher than unity while the intercepts were not significantly different than zero, indicating that underestimation of outdoor $PM_{2.5}$ concentrations at School B would result if concentrations at these sites were used as surrogates.

In the summer, variability in $PM_{2.5}$ concentrations at FSMs were weak predictors of variability in transect concentrations at both schools, with R^2 less than 0.20 for School A, and R^2 less than 0.25 for School B. For both schools and all FSMs, the intercepts of LRMs were significantly higher than zero, indicating spatial heterogeneity in $PM_{2.5}$ concentrations due to the impact of local emissions. Some of the $PF_{s,FSM,s}$ estimates were insignificant, such as at FSMs R1, R2, G1, G6, G8, and G9 for School A and FSMs R1, R3, G1−4, G6, G8, and G9 for School B, which implies that FSMs are not useful as surrogates for selected schools in summer when local pollution dominates.

Exposure Error. Errors in hourly indoor exposure estimates using different outdoor surrogates are summarized as boxplots by school and season in Figure 3. In winter, the errors in estimated indoor exposure based on using outdoor concentrations measured at nearby transects (Scenario S1) were small, with mean errors not exceeding 2% for both schools and interquartile ranges of errors not exceeding ±6%. The narrow range of errors indicates a high level of confidence in indoor exposure estimates developed using nearby outdoor concentrations when the infiltration factor is high.

The exposure errors introduced from using outdoor concentrations at FSMs in Scenario S2 were much wider compared to Scenario S1 in winter, with interquartile ranges of relative exposure error approximately 2−4 times wider than those under S1 for both schools. The mean exposure error under S2 varied substantially among FSM sites, ranging from −23% to 29% for School A and from −30% to 11% for School B.

The exposure errors introduced from using PF-corrected outdoor concentrations at FSMs (Scenario S3) were substantially lower than those under Scenario S2 in winter. For example, the mean error of estimated hourly indoor concentration at School A was reduced from −23% under S2

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to 6% under S3 at G6, and from −21% under S2 to 4% under S3 at G8. The mean error for School B changed from −28% to 6% under S2 to less than ±3% for the five sites with $PF_{B,FSM,W}$ significantly higher than unity. The interquartile ranges of exposure error under S3 were similar to those under Scenario S2 in winter. Thus, $PF_{s,FSM,W}$ are useful in correcting for bias but may not improve the precision of exposure concentration estimates.

In summer, the mean error under S1 was 12% for School A and 14% for School B, with interquartile ranges from −16% to 29% and from −15% to 38%, respectively. Positive mean errors were observed for both schools at all FSMs, indicating an overestimation of exposure since $PF_{s,FSM,S}$ were all less than unity. The mean exposure error introduced from using FSMs as surrogates was reduced from up to 88% and 92% under S2 for School A and B, respectively, to less than 11% under S3 for both schools after $PF_{s,FSM,S}$ correction. This is consistent with the findings in winter that PF is useful in correcting for the bias in exposure concentrations estimates if FSMs are used as outdoor surrogates.

The summer mean exposure errors were much larger and the range of errors was wider compared to those in winter. However, the indoor concentrations in summer were generally below 10 $\mu g/m^3$. The ratio of average indoor $PM_{2.5}$ concentrations in winter to summer was 7 for school A and 12 for school B. Therefore, the exposure errors in summer are not expected to substantially affect the annual exposure to $PM_{2.5}$ at both schools.

Concordance Indices. A selection of 19 potential concordance indices were evaluated based on correlation with $PF_{s,FSM,p}$ values in Table 1. Among the single parameter concordance indices, significant correlations were found between $\Delta W_{s,FSM}$ and $PF_{s,FSM,p}$, with SCC of 0.59 for School A and 0.71 for School B, respectively, among all FSMs and seasons. This indicates that wind direction is the major factor affecting the spatial variability in $PM_{2.5}$ concentrations between selected schools and FSMs.

The correlation with $PF_{s,FSM,p}$ was low and not statistically significant for $D_{s,FSM}$, $\Delta E_{s,FSM}$, $\Delta H_{s,FSM}$, and their related permutations, such as inverse and log form. However, these CIs may interact with $\Delta W_{s,FSM}$ and affect correlations with $PF_{s,FSM,p}$, as listed in Table 1. Among the mixed CIs, statistically significant correlations were found for both schools between $PF_{s,FSM,p}$ and the product of logarithmic $D_{s,FSM}$ and $\Delta W_{s,FSM}$ (MIX1), the product of logarithmic $\Delta H_{s,FSM}$ and $\Delta W_{s,FSM}$ (MIX3), and the product of logarithmic $D_{s,FSM}$, logarithmic $\Delta H_{s,FSM}$ and $\Delta W_{s,FSM}$ (MIX5). This indicates that variability in PF values are mainly affected by the direction to the school relative to the air pollution flow ($\Delta W_{s,FSM}$) and marginally affected by the spatial differences in geographic locations ($D_{s,FSM}$), and proximity to ground emission sources ($\Delta H_{s,FSM}$).

The final model developed for $PF_{s,FSM}$ is given in Table 1, with $\Delta W_{s,FSM}$ and the product of logarithmic $D_{s,FSM}$, logarithmic $\Delta H_{s,FSM}$ and $\Delta W_{s,FSM}$ (MIX5) as independent variables. The final model explained 53% (R^2 = 0.53) of the variation in $PF_{s,FSM}$. Wind direction is the major predictor of $PF_{s,FSM}$. Shifts in the dominant wind direction under Asian Monsoon system significantly changed the direction of air flow and the abundance of inflow of PM pollution from Asia to Hong Kong. Spatial distance and proximity to emission source also affect the magnitude of $PF_{s,FSM}$, which is consistent with the association analysis.

DISCUSSION

Variations in outdoor $PM_{2.5}$ concentrations were strong predictors of variations in indoor exposure to $PM_{2.5}$ at both schools in winter (R^2 > 0.98), when natural ventilation was used, but weak indicators in summer (R^2 < 0.13), when A/C was used. The use of A/C is usually in conjunction with closed windows, which reduces the air exchange and thus reduces the infiltration of outdoor particles. Seasonal variations in health risks related to $PM_{2.5}$ exposures have been widely observed in epidemiological studies in which ambient $PM_{2.5}$ concentrations were used as surrogates.[14−17,75,76] These seasonal variations in the health estimates are in part because of the differences in infiltration of outdoor particles due to the changes in ventilation operation. For example, Franklin et al. found that increased prevalence of central air conditioning was associated with a decreased effect of $PM_{2.5}$ regarding all-cause and specific-cause mortality in 27 US communities.[15]

Another factor that may contribute to the seasonal variations observed in epidemiological studies is seasonal differences in the spatial distribution of outdoor $PM_{2.5}$ concentrations.[14−18] The exposure errors introduced by using FSMs as surrogates in summer were found to be an average of 3−5 times higher than those in winter. These seasonal variations in exposure error could contribute to the decreased magnitude of effects estimates and lower statistical power in epidemiological studies. For example, Ko et al. found that the warm season (summer) was associated with a lower risk of hospital admission for asthma compared with the cold season (winter) in Hong Kong, given the same 10 $\mu g/m^3$ change in ambient $PM_{2.5}$ concentrations.[77]

PF was found to be useful in correcting the systematic positive or negative exposure errors introduced by using FSM as surrogates. The mean errors in hourly $PM_{2.5}$ exposures were reduced from up to 92% to less than 20% for all schools and seasons after taking into account the seasonal-specific PF values (Scenario S3). The case study showed that magnitude of the PF values can be inferred or estimated for Hong Kong based on a few geographic and meteorological parameters, as listed in Table 1. This demonstrates a way to estimate and adjust for seasonal variations in exposure and, thus, the seasonal variations observed in the epidemiological studies. The uncertainty of the PF values can be estimated based on the standard error of the parameters in the final model in Table 1, and can be further used to estimate the exposure uncertainty.

In microenvironmental models, PF is usually regarded as a static correction factor that only depends on distance between FSM and the targeted microenvironment.[27] However, PF is affected by the direction of wind which determines the direction of air flow and pollution transport. The wind direction changes with time. Therefore, variations in dominant wind direction should be taken into account in developing PFs to address temporal differences in the spatial distribution of outdoor $PM_{2.5}$ concentrations. Variability in PF values was found to be associated with variability in the dominant wind direction, GIS-related parameters such as distance and elevation differences between schools and FSMs, and differences in sampling height. Land use type has been used to explain the spatial variability in outdoor $PM_{2.5}$ in land use regression models.[26] However, the impact of land use on PF could not be assessed because the two schools and most of the

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FSMs are located in urbanized areas. Future work is needed to assess whether PFs are sensitive to land use.

The model developed for PF reflects the key sources of spatial variations for $PM_{2.5}$ pollution, such as the dominant wind direction and proximity to ground emissions. However, the numeric values may vary from city to city. It is recommended to conduct replicate studies in other cities to verify the results and develop city-specific models between PF and concordance indices for better use in exposure and epidemiological studies. Nevertheless, the recommendations to use fixed sites located along the prevailing wind direction and in close proximity are expected to be robust. In cases where ambient data from multiple FSMs are available, the best choice of a preferred FSM should not be based only on the shortest distance but should also take into account which FSM is located along the prevailing direction of air pollution flow, as well as the proximity to ground emission.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b04474.

Description of selected schools and FSMs is listed in Table S1. A summary for measurement at selected schools are given in Table S2. Hourly average $PM_{2.5}$concentrations at schools and fixed site monitors (FSMs) are listed in Table S3. Spearman Correlation Coefficients (SCC) between indoor and outdoor $PM_{2.5}$ concentrations and between school outdoor measurements and FSMs are listed in Table S4 and S5, respectively. Infiltration factors estimated with and without autocorrelation are listed in Table S6. Proximity factors estimated between selected schools and FSMs are listed in Table S7. The instrument package and results of instrument calibration are shown in Figure S1 and S2, respectively. Examples of time series of measured $PM_{2.5}$ concentrations at selected schools during winter and summer are shown in Figure S3. Wind rose maps from a background meteorological station in Hong Kong during winter and summer sampling periods are shown in Figure S4 (PDF)

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Notes

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■ REFERENCES

(1) *Integrated Science Assessment for Particulate Matter*. In U.S. Environmental Protection Agency: Research Triangle Park, NC, 2009.

(2) Eftim, S. E.; Samet, J. M.; Janes, H.; McDermott, A.; Dominici, F. Fine particulate matter and mortality - A comparison of the six cities and American Cancer Society cohorts with a medicare cohort. *Epidemiology* **2008**, *19* (2), 209−216.

(3) Zeger, S. L.; Dominici, F.; McDermott, A.; Samet, J. M. Mortality in the Medicare Population and Chronic Exposure to Fine Particulate Air Pollution in Urban Centers (2000−2005). *Environ. Health Perspect.* **2008**, *116* (12), 1614−1619.

(4) Miller, K. A.; Siscovick, D. S.; Sheppard, L.; Shepherd, K.; Sullivan, J. H.; Anderson, G. L.; Kaufman, J. D. Long-term exposure to air pollution and incidence of cardiovascular events in women. *N. Engl. J. Med.* **2007**, *356* (5), 447−458.

(5) Pope, C. A.; Dockery, D. W. Health effects of fine particulate air pollution: Lines that connect. *J. Air Waste Manage. Assoc.* **2006**, *56* (6), 709−742.

(6) Zanobetti, A.; Schwartz, J. The Effect of Fine and Coarse Particulate Air Pollution on Mortality: A National Analysis. *Environ. Health Perspect.* **2009**, *117* (6), 898−903.

(7) Dominici, F.; Peng, R. D.; Bell, M. L.; Pham, L.; McDermott, A.; Zeger, S. L.; Samet, J. M. Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases. *Jama-J. Am. Med. Assoc* **2006**, *295* (10), 1127−1134.

(8) Pope, C. A.; Renlund, D. G.; Kfoury, A. G.; May, H. T.; Horne, B. D. Relation of Heart Failure Hospitalization to Exposure to Fine Particulate Air Pollution. *Am. J. Cardiol.* **2008**, *102* (9), 1230−1234.

(9) Rich, D. Q.; Schwartz, J.; Mittleman, M. A.; Link, M.; Luttmann-Gibson, H.; Catalano, P. J.; Speizer, F. E.; Dockery, D. W. Association of short-term ambient air pollution concentrations and ventricular arrhythmias. *Am. J. Epidemiol.* **2005**, *161* (12), 1123−1132.

(10) *Ambient Air Pollution: A Global Assessment of Exposure and Burden of Disease*, 9789241511353; World Health Organization: Gevena, Swizerland, 2016.

(11) Wilson, W. E.; Brauer, M. Estimation of ambient and non-ambient components of particulate matter exposure from a personal monitoring panel study. *J. Exposure Sci. Environ. Epidemiol.* **2006**, *16* (3), 264−274.

(12) Burke, J. M.; Zufall, M. J.; Ozkaynak, H. A population exposure model for particulate matter: case study results for $PM_{2.5}$ in Philadelphia, PA. *J. Exposure Sci. Environ. Epidemiol.* **2001**, *11* (6), 470−489.

(13) *Guidelines for Exposure Assessment*. In U.S. Environmental Protection Agency: Washington, D.C., 1992; Vol. *57*, pp 22888−22938.

(14) Ostro, B.; Feng, W. Y.; Broadwin, R.; Green, S.; Lipsett, M. The effects of components of fine particulate air pollution on mortality in California: Results from CALFINE. *Environ. Health Perspect.* **2007**, *115* (1), 13−19.

(15) Franklin, M.; Zeka, A.; Schwartz, J. Association between $PM_{2.5}$ and all-cause and specific-cause mortality in 27 US communities. *J. Exposure Sci. Environ. Epidemiol.* **2007**, *17* (3), 279−87.

(16) Bell, M. L.; Ebisu, K.; Peng, R. D.; Walker, J.; Samet, J. M.; Zeger, S. L.; Dominici, F. Seasonal and regional short-term effects of fine particles on hospital admissions in 202 US counties, 1999−2005. *Am. J. Epidemiol.* **2008**, *168* (11), 1301−10.

(17) Pascal, M.; Falq, G.; Wagner, V.; Chatignoux, E.; Corso, M.; Blanchard, M.; Host, S.; Pascal, L.; Larrieu, S. Short-term impacts of particulate matter (PM_{10}, $PM_{10-2.5}$, $PM_{2.5}$) on mortality in nine French cities. *Atmos. Environ.* **2014**, *95*, 175−184.

(18) Vrijheid, M.; Martinez, D.; Manzanares, S.; Dadvand, P.; Schembari, A.; Rankin, J.; Nieuwenhuijsen, M. Ambient Air Pollution and Risk of Congenital Anomalies: A Systematic Review and Meta-analysis. *Environ. Health Perspect.* **2011**, *119* (5), 598−606.

(19) Dionisio, K. L.; Baxter, L. K.; Chang, H. H. An Empirical Assessment of Exposure Measurement Error and Effect Attenuation in Bipollutant Epidemiologic Models. *Environ. Health Perspect.* **2014**, *122* (11), 1216−1224.

(20) Sarnat, S. E.; Klein, M.; Sarnat, J. A.; Flanders, W. D.; Waller, L. A.; Mulholland, J. A.; Russell, A. G.; Tolbert, P. E. An examination of exposure measurement error from air pollutant spatial variability in time-series studies. *J. Exposure Sci. Environ. Epidemiol.* **2010**, *20* (2), 135−146.
(21) Zeger, S. L.; Thomas, D.; Dominici, F.; Samet, J. M.; Schwartz, J.; Dockery, D.; Cohen, A. Exposure measurement error in time-series studies of air pollution: concepts and consequences. *Environ. Health Perspect.* **2000**, *108* (5), 419−426.
(22) Ashmore, M. R.; Dimitroulopoulou, C. Personal exposure of children to air pollution. *Atmos. Environ.* **2009**, *43* (1), 128−141.
(23) Meng, Q. Y.; Turpin, B. J.; Korn, L.; Weisel, C. P.; Morandi, M.; Colome, S.; Zhang, J. F. J.; Stock, T.; Spektor, D.; Winer, A.; Zhang, L.; Lee, J. H.; Giovanetti, R.; Cui, W.; Kwon, J.; Alimokhtari, S.; Shendell, D.; Jones, J.; Farrar, C.; Maberti, S. Influence of ambient (outdoor) sources on residential indoor and personal PM2.5 concentrations: Analyses of RIOPA data. *J. Exposure Sci. Environ. Epidemiol.* **2005**, *15* (1), 17−28.
(24) Hanninen, O. O.; Lebret, E.; Ilacqua, V.; Katsouyanni, K.; Kunzli, F.; Sram, R. J.; Jantunen, M. Infiltration of ambient $PM_{2.5}$ and levels of indoor generated non-ETS $PM_{2.5}$ in residences of four European cities. *Atmos. Environ.* **2004**, *38* (37), 6411−6423.
(25) Hodas, N.; Meng, Q. Y.; Lunden, M. M.; Rich, D. Q.; Ozkaynak, H.; Baxter, L. K.; Zhang, Q.; Turpin, B. J. Variability in the fraction of ambient fine particulate matter found indoors and observed heterogeneity in health effect estimates. *J. Exposure Sci. Environ. Epidemiol.* **2012**, *22* (5), 448−454.
(26) Hoek, G.; Beelen, R.; de Hoogh, K.; Vienneau, D.; Gulliver, J.; Fischer, P.; Briggs, D. A review of land-use regression models to assess spatial variation of outdoor air pollution. *Atmos. Environ.* **2008**, *42* (33), 7561−7578.
(27) *Air Pollutants Exposure Model Documentation* (APEX, Version 5) Vol. II, EPA-452/R-17-001b; Technical Support Document: Research Triangle Park, NC, 2017.
(28) Che, W.; Frey, H. C.; Lau, A. K. Comparison of Sources of Variability in School Age Children Exposure to Ambient $PM_{2.5}$. *Environ. Sci. Technol.* **2015**, *49* (3), 1511−1520.
(29) Johnson, T. R.; Langstaff, J. E.; Graham, S.; Fujita, E. M.; Campbell, D. E. A multipollutant evaluation of APEX using microenvironmental ozone, carbon monoxide, and particulate matter ($PM_{2.5}$) concentrations measured in Los Angeles by the exposure classification project. *Cogent Environmental Science* **2018**, *4* (1), 1453022.
(30) Wang, S.; Li, G. G.; Gong, Z. Y.; Du, L.; Zhou, Q. T.; Meng, X. Y.; Xie, S. Y.; Zhou, L. Spatial distribution, seasonal variation and regionalization of $PM_{2.5}$ concentrations in China. *Sci. China: Chem.* **2015**, *58* (9), 1435−1443.
(31) Xie, M. J.; Coons, T. L.; Dutton, S. J.; Milford, J. B.; Miller, S. L.; Peel, J. L.; Vedal, S.; Hannigan, M. P. Intra-urban spatial variability of $PM_{2.5}$-bound carbonaceous components. *Atmos. Environ.* **2012**, *60*, 486−494.
(32) Masiol, M.; Benetello, F.; Harrison, R. M.; Formenton, G.; De Gaspari, F.; Pavoni, B. Spatial, seasonal trends and transboundary transport of $PM_{2.5}$ inorganic ions in the Veneto region (Northeastern Italy). *Atmos. Environ.* **2015**, *117*, 19−31.
(33) Raysoni, A. U.; Sarnat, J. A.; Sarnat, S. E.; Garcia, J. H.; Holguin, F.; Luevano, S. F.; Li, W. W. Binational school-based monitoring of traffic-related air pollutants in El Paso, Texas (USA) and Ciudad Juarez, Chihuahua (Mexico). *Environ. Pollut.* **2011**, *159* (10), 2476−86.
(34) Nuvolone, D.; della Maggiore, R.; Maio, S.; Fresco, R.; Baldacci, S.; Carrozzi, L.; Pistelli, F.; Viegi, G. Geographical information system and environmental epidemiology: a cross-sectional spatial analysis of the effects of traffic-related air pollution on population respiratory health. *Environ. Health* **2011**, *10* (1), 1−12.
(35) Eeftens, M.; Beelen, R.; de Hoogh, K.; Bellander, T.; Cesaroni, G.; Cirach, M.; Declercq, C.; Dedele, A.; Dons, E.; de Nazelle, A.; Dimakopoulou, K.; Eriksen, K.; Falq, G.; Fischer, P.; Galassi, C.; Grazuleviciene, R.; Heinrich, J.; Hoffmann, B.; Jerrett, M.; Keidel, D.; Korek, M.; Lanki, T.; Lindley, S.; Madsen, C.; Molter, A.; Nador, G.; Nieuwenhuijsen, M.; Nonnemacher, M.; Pedeli, X.; Raaschou-Nielsen, O.; Patelarou, E.; Quass, U.; Ranzi, A.; Schindler, C.; Stempfelet, M.; Stephanou, E.; Sugiri, D.; Tsai, M. Y.; Yli-Tuomi, T.; Varro, M. J.; Vienneau, D.; von Klot, S.; Wolf, K.; Brunekreef, B.; Hoek, G. Development of Land Use Regression Models for $PM_{2.5}$, $PM_{2.5}$ Absorbance, PM_{10} and PMcoarse in 20 European Study Areas; Results of the ESCAPE Project. *Environ. Sci. Technol.* **2012**, *46* (20), 11195−11205.
(36) Liu, Y.; Paciorek, C. J.; Koutrakis, P. Estimating Regional Spatial and Temporal Variability of $PM_{2.5}$ Concentrations Using Satellite Data, Meteorology, and Land Use Information. *Environ. Health Perspect.* **2009**, *117* (6), 886−892.
(37) Zhai, L.; Zou, B.; Fang, X.; Luo, Y. Q.; Wan, N.; Li, S. Land Use Regression Modeling of $PM_{2.5}$ Concentrations at Optimized Spatial Scales. *Atmosphere* **2017**, *8* (1), 1−15.
(38) Shi, Y.; Lau, K. K. L.; Ng, E. Developing Street-Level $PM_{2.5}$ and PM_{10} Land Use Regression Models in High-Density Hong Kong with Urban Morphological Factors. *Environ. Sci. Technol.* **2016**, *50* (15), 8178−8187.
(39) Guo, H.; Ding, A. J.; So, K. L.; Ayoko, G.; Li, Y. S.; Hung, W. T. Receptor modeling of source apportionment of Hong Kong aerosols and the implication of urban and regional contribution. *Atmos. Environ.* **2009**, *43* (6), 1159−1169.
(40) Arain, M. A.; Blair, R.; Finkelstein, N.; Brook, J. R.; Sahsuvaroglu, T.; Beckerman, B.; Zhang, L.; Jerrett, M. The use of wind fields in a land use regression model to predict air pollution concentrations for health exposure studies. *Atmos. Environ.* **2007**, *41* (16), 3453−3464.
(41) Guerra, S. A.; Lane, D. D.; Marotz, G. A.; Carter, R. E.; Hohl, C. M.; Baldauf, R. W. Effects of wind direction on coarse and fine particulate matter concentrations in southeast Kansas. *J. Air Waste Manage. Assoc.* **2006**, *56* (11), 1525−1531.
(42) Yousaf, A. R.; Khan, N. The study of particulate matter concentration in schools of Lahore. *Nature Environment and Pollution Technology* **2013**, *12* (2), 289−296.
(43) Altug, H.; Gaga, E. O.; Dogeroglu, T.; Brunekreef, B.; Hoek, G.; Van Doorn, W. Effects of ambient air pollution on respiratory tract complaints and airway inflammation in primary school children. *Sci. Total Environ.* **2014**, *479*−*480*, 201−9.
(44) Annesi-Maesano, I.; Moreau, D.; Caillaud, D.; Lavaud, F.; Le Moullec, Y.; Taytard, A.; Pauli, G.; Charpin, D. Residential proximity fine particles related to allergic sensitisation and asthma in primary school children. *Respiratory Medicine* **2007**, *101* (8), 1721−1729.
(45) Ceretti, E.; Feretti, D.; Viola, G. C. V.; Zerbini, I.; Limina, R. M.; Zani, C.; Capelli, M.; Lamera, R.; Donato, F.; Gelatti, U. DNA damage in buccal mucosa cells of pre-school children exposed to high levels of urban air pollutants. *PLoS One* **2014**, *9* (5), 1−9.
(46) Epton, M. J.; Dawson, R. D.; Brooks, W. M.; Kingham, S.; Aberkane, T.; Cavanagh, J. A.; Frampton, C. M.; Hewitt, T.; Cook, J. M.; McLeod, S.; McCartin, F.; Trought, K.; Brown, L. The effect of ambient air pollution on respiratory health of school children: a panel study. *Environ. Health* **2008**, *7* (16), 1−11.
(47) Millstein, J.; Gilliland, F.; Berhane, K.; Gauderman, W. J.; McConnell, R.; Avol, E.; Rappaport, E. B.; Peters, J. M. Effects of ambient air pollutants on asthma medication use and wheezing among fourth-grade school children from 12 southern California communities enrolled in the children's health study. *Arch. Environ. Health* **2004**, *59* (10), 505−514.
(48) Silcox, G. D.; Kelly, K. E.; Crosman, E. T.; Whiteman, C. D.; Allen, B. L. Wintertime $PM_{2.5}$ concentrations during persistent, multi-day cold-air pools in a mountain valley. *Atmos. Environ.* **2012**, *46*, 17−24.
(49) So, K. L.; Guo, H.; Li, Y. S. Long-term variation of $PM_{2.5}$ levels and composition at rural, urban, and roadside sites in Hong Kong: Increasing impact of regional air pollution. *Atmos. Environ.* **2007**, *41* (40), 9427−9434.
(50) Louie, P. K. K.; Chow, J. C.; Chen, L. W. A.; Watson, J. G.; Leung, G.; Sin, D. W. M. $PM_{2.5}$ chemical composition in Hong Kong:

DOI: 10.1021/acs.est.8b04474
Environ. Sci. Technol. 2019, 53, 808−819

Urban and regional variations. *Sci. Total Environ.* **2005**, *338* (3), 267−281.
(51) Wang, T.; Guo, H.; Blake, D. R.; Kwok, Y. H.; Simpson, I. J.; Li, Y. S. Measurements of trace gases in the inflow of South China Sea background air and outflow of regional pollution at Tai O, Southern China. *J. Atmos. Chem.* **2005**, *52* (3), 295−317.
(52) Guo, H.; Morawska, L.; He, C.; Gilbert, D. Impact of ventilation scenario on air exchange rates and on indoor particle number concentrations in an air-conditioned classroom. *Atmos. Environ.* **2008**, *42* (4), 757−768.
(53) Heudorf, U.; Neitzert, V.; Spark, J. Particulate matter and carbon dioxide in classrooms - The impact of cleaning and ventilation. *Int. J. Hyg. Environ. Health* **2009**, *212* (1), 45−55.
(54) Dorizas, P. V.; Assimakopoulos, M. N.; Helmis, C.; Santamouris, M. An integrated evaluation study of the ventilation rate, the exposure and the indoor air quality in naturally ventilated classrooms in the Mediterranean region during spring. *Sci. Total Environ.* **2015**, *502*, 557−570.
(55) *A Supplement on Ventilation - Guidelines on Prevention of Communicable Diseases in Schools/ Kindergartens/Kindergartens Cum Child Care-Centres/ Child Care Centres*; Center for Health Protection, Department of Health: Hong Kong, 2014.
(56) *Hong Kong Annual Digest of Statistics*. In Census and Statistics Department: Hong Kong, 2017.
(57) *Hong Kong: The Facts*; Information Services Department: Hong Kong, 2017.
(58) *Food Safety of School Lunchboxes*. https://www.cfs.gov.hk/english/programme/programme_haccp/programme_haccp_lunchbox_school.html (accessed on August 10 2018).
(59) *The Annual Traffic Census 2016*. In Transport Department: Hong Kong, 2017.
(60) Kim, J. Y.; Magari, S. R.; Herrick, R. F.; Smith, T. J.; Christiani, D. C. Comparison of fine particle measurements from a direct-reading instrument and a gravimetric sampling method. *J. Occup. Environ. Hyg.* **2004**, *1* (11), 707−715.
(61) Yanosky, J. D.; Williams, P. L.; MacIntosh, D. L. A comparison of two direct-reading aerosol monitors with the federal reference method for $PM_{2.5}$ in indoor air. *Atmos. Environ.* **2002**, *36* (1), 107−113.
(62) Edimansyah, B. A.; Rusli, B. N.; Naing, L.; Azwan, B. A.; Aziah, B. D. Indoor air quality in an automotive assembly plant in Selangor, Malaysia. *Southeast Asian J. Trop. Med. Public Health* **2009**, *40* (1), 187−192.
(63) Apte, M. G.; Fisk, W. J.; Daisey, J. M. Associations between indoor CO_2 concentrations and sick building syndrome symptoms in US office buildings: An analysis of the 1994−1996 BASE study data. *Indoor Air* **2000**, *10* (4), 246−257.
(64) Chung, A.; Chang, D. P. Y.; Kleeman, M. J.; Perry, K. D.; Cahill, T. A.; Dutcher, D.; McDougall, E. M.; Stroud, K. Comparison of real-time instruments used to monitor airborne particulate matter. *J. Air Waste Manage. Assoc.* **2001**, *51* (1), 109−120.
(65) Che, W. W.; Frey, H. C.; Lau, A. K. H. Sequential Measurement of Intermodal Variability in Public Transportation $PM_{2.5}$ and CO Exposure Concentrations. *Environ. Sci. Technol.* **2016**, *50* (16), 8760−8769.
(66) Csavina, J.; Field, J.; Felix, O.; Corral-Avitia, A. Y.; Saez, A. E.; Betterton, E. A. Effect of wind speed and relative humidity on atmospheric dust concentrations in semi-arid climates. *Sci. Total Environ.* **2014**, *487*, 82−90.
(67) Wilson, J. C. Aerosol Technology - Properties, Behavior, and Measurement of Airborne Particles - Hinds,Wc. *Am. Sci.* **1983**, *71* (4), 430−431.
(68) *List of Designated Reference and Equivalent Methods*; United States Environmental Protection Agency: Research Triangle Park, NC, 2016.
(69) Tam, W. S. *A Report on the Results From the Air Quality Monitoring Network (AQMN)*, (2016); EPD/TR 1/17; Hong Kong, 2017.
(70) Hamby, D. M. The Gaussian atmospheric transport model and its sensitivity to the joint frequency distribution and parametric variability. *Health Phys.* **2002**, *82* (1), 64−73.
(71) Lushi, E.; Stockie, J. M. An inverse Gaussian plume approach for estimating atmospheric pollutant emissions from multiple point sources. *Atmos. Environ.* **2010**, *44* (8), 1097−1107.
(72) Mukaka, M. M. Statistics Corner: A guide to appropriate use of Correlation coefficient in medical research. *Malawi Med. J.* **2012**, *24* (3), 69−71.
(73) Levy, J. I.; Dumyahn, T.; Spengler, J. D. Particulate matter and polycyclic aromatic hydrocarbon concentrations in indoor and outdoor microenvironments in Boston, Massachusetts. *J. Exposure Sci. Environ. Epidemiol.* **2002**, *12* (2), 104−114.
(74) Houseman, E. A.; Ryan, L.; Levy, J. I.; Spengler, J. D. Autocorrelation in real-time continuous monitoring of microenvironments. *J. Appl. Stat* **2002**, *29* (6), 855−872.
(75) Ito, K.; Mathes, R.; Ross, Z.; Nadas, A.; Thurston, G.; Matte, T. Fine Particulate Matter Constituents Associated with Cardiovascular Hospitalizations and Mortality in New York City. *Environ. Health Perspect.* **2011**, *119* (4), 467−473.
(76) Zhou, J. A.; Ito, K.; Lall, R.; Lippmann, M.; Thurston, G. Time-Series Analysis of Mortality Effects of Fine Particulate Matter Components in Detroit and Seattle. *Environ. Health Perspect.* **2011**, *119* (4), 461−466.
(77) Ko, F. W. S.; Tam, W.; Wong, T. W.; Lai, C. K. W.; Wong, G. W. K.; Leung, T. F.; Ng, S. S. S.; Hui, D. S. C. Effects of air pollution on asthma hospitalization rates in different age groups in Hong Kong. *Clin. Exp. Allergy* **2007**, *37* (9), 1312−1319.

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Environ. Sci. Technol. 2019, 53, 808−819