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Evaluation of Variability in the Ambient $PM_{2.5}$ Concentrations from FEM and FRM-like Measurements for Exposure Estimates

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ABSTRACT

This study aims to evaluate the variability in ambient fine particulate matter ($PM_{2.5}$) concentrations obtained from the federal equivalent method (FEM) and federal reference method (FRM)-like measurements at the national air quality monitoring stations (AQMSs) for exposure estimates and to examine the effect of environmental factors and sampling site characteristics affecting the spatial and temporal variations in $PM_{2.5}$ concentrations. A mixed-effects model was used to evaluate the temporal and spatial variability in daily and annual $PM_{2.5}$ concentrations during 2014–2017 at 16 AQMSs in the Big Taipei City, Taiwan. The mean FEM $PM_{2.5}$ concentrations were ~30% higher than the FRM-like $PM_{2.5}$ concentrations. The FRM-like $PM_{2.5}$ concentrations obtained by applying the calibration procedures presented a negligible between-site variability. The daily FEM $PM_{2.5}$ concentrations were dominated by the within-site variability (~90%), whereas the annual concentrations were reasonably attributable to the between-site variability (47.8%). Ambient $PM_{2.5}$ was mainly affected by the gaseous pollutants (such as NO_2, O_3, and SO_2), accounting for 45.8% and 26.8% of the within-site and between-site variability in concentrations, respectively. The FEM measurements rather than the FRM-like measurements at the AQMSs could provide a higher between-site variability for exposure estimates of $PM_{2.5}$ in the epidemiological studies.

Keywords: Ambient $PM_{2.5}$, Calibration, Within and between variability, Exposure estimates

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1 INTRODUCTION

The elevated levels of fine particulate matter ($PM_{2.5}$) are associated with adverse health effects, such as respiratory and cardiovascular morbidity and mortality, which have been reported in many studies (Beelen *et al.*, 2014; Cai *et al.*, 2016; Kaufman *et al.*, 2016). The World Health Organization (WHO, 2014) has indicated that outdoor $PM_{2.5}$ accounts for 7 million deaths worldwide every year. Recent literature has documented that exposure to even low levels of $PM_{2.5}$ can significantly increase all-cause mortality (Shi *et al.*, 2016). National ambient air quality standards (NAAQS) are usually regulated in many countries to limit air pollution (such as ambient $PM_{2.5}$ levels) and protect public health. For instance, the annual (15 $\mu g\ m^{-3}$) and daily (35 $\mu g\ m^{-3}$) standards of $PM_{2.5}$ have been set by Taiwan EPA while lower guideline limits for outdoor $PM_{2.5}$ (annual = 10 $\mu g\ m^{-3}$, daily = 25 $\mu g\ m^{-3}$) have been suggested by WHO. The filter-based instruments such as BGI PQ200 and RAAS2.5 single and multi-day samplers are usually used for consistent and repeatable measurements of 24-h $PM_{2.5}$ concentrations following the federal reference method (FRM) certified by USEPA (McNamara *et al.*, 2011; Kelly *et al.*, 2017). In addition, the continuous monitoring measurements following the federal equivalent method (FEM) or non-FRM based on optical, beta ray attenuation, or tapered element oscillation microbalance (TEOM) monitoring have also been widely used for timely $PM_{2.5}$ measurements in the metropolitan area. In Taiwan, hourly concentrations of FEM $PM_{2.5}$ from 76 air quality monitoring stations (AQMSs) of Taiwan EPA (https://data.epa.gov.tw/dataset/aqx_p_15) have been announced since 2006. FEM $PM_{2.5}$ concentrations obtained via optical measurements are usually affected by humidity, temperature, size distribution, and chemical composition (Bortnick *et al.*, 2002; Dinoi *et al.*, 2017; Sofowote *et al.*, 2014). Thus, the calibration of the FEM $PM_{2.5}$ data with the FRM $PM_{2.5}$ measurements through a statistical linear regression model (so-called FRM-like $PM_{2.5}$) is necessary. Bortnick *et al.* (2002) indicated that the resulting FRM-like $PM_{2.5}$ measurements using a linear regression model were able to provide more timely reporting of $PM_{2.5}$. The calibration approach of the data quality objective (DQO) process consists of a seven-step strategy, as suggested by U.S. EPA (U.S. EPA, 1994).

The FRM-like measurements of $PM_{2.5}$ are performed to not only provide a comprehensive assessment of air quality for the public but also to investigate its impact on human health. Numerous population-based epidemiological studies of air pollution usually rely on FRM-like $PM_{2.5}$ measurements either by direct methods or using geographical modeling, i.e., microenvironmental exposure, land use regression (LUR), and kriging models to estimate $PM_{2.5}$ exposure (Koenig *et al.*, 2005; Meng *et al.*, 2005; Laden *et al.*, 2006; Krewski *et al.*, 2009; Jerrett *et al.*, 2013; Özkaynak *et al.*, 2013; Kioumourtzoglou *et al.*, 2014). However, it is not clear whether the $PM_{2.5}$ estimates from FRM-like measurements through a calibration process attenuate exposure variability or lead to non-differential misclassification of exposure for population-based health studies. For instance, a model could result in misleading $PM_{2.5}$ concentrations, when it is mainly affected by meteorological conditions such as temperature (Bortnick *et al.*, 2002). The exposure error obtained from ambient $PM_{2.5}$ measurements can impact observed health risks, potentially distorting associations and interactions between covariates and outcomes, and leading to invalid inferences. To date, no study has explored the applicability of uncalibrated FEM and calibrated FRM-like measurements of $PM_{2.5}$ for exposure estimates in health effect studies. On the other hand, a large number of studies have examined the effect of meteorological conditions and gaseous pollutants on $PM_{2.5}$ concentrations (Ito *et al.*, 2007; Zhang *et al.*, 2015). These potential confounders, such as temperature, NO_2, and O_3, are usually included for adjustment in the health effect models (Crouse *et al.*, 2015; Luo *et al.*, 2016). Thus, it is important to understand the magnitude of the impact of these factors on within- and between-site (or -group) variability in concentrations at different time intervals. This information is vital for the future design of studies to improve exposure estimates associated with mortality and morbidity outcomes.

This study aims to evaluate $PM_{2.5}$ variability in FRM-like and FEM measurements and identify the important factors affecting within- and between-site (-group) variability in $PM_{2.5}$ concentrations in the metropolitan area. This study, a part of the Taiwan Health and Air Pollution study (THAP), can make an effort on the improvement of exposure estimates for population-based health risk analysis.

2 METHODS

2.1 Data Sources and Calibration Process

In this study, we selected Taipei and New Taipei cities, (together known as Big Taipei City), having a population of 7 million in 2,225 km^2 of land, as our study area because it has a high-density of national AQMSs. There are 19 national AQMSs (Fig. 1), including one national park AQMS (Yangming mountain), three traffic AQMS (Sanchong, Yonghe, and Datong), one background AQMS (FugueiCape), and 14 general AQMSs (Tu-cheng (TC), Shi-lin (SL), Zhong-shan (ZS), Gu-ting (GT), Xi-zhi (XiZ), Song-shan (SS), Ban-qiao (BQ), Lin-kou (LK), Tam-sui (TS), Cai-liao (CL), Xin-dian (XD), Xin-zhuang (XinZ), Wanli (WL), and Wan-hua (WH)). Out of these 14 general stations, one is the background station (WL) for the specific purpose of air quality monitoring. After excluding national park and background monitoring stations, the air quality monitoring data from a total of

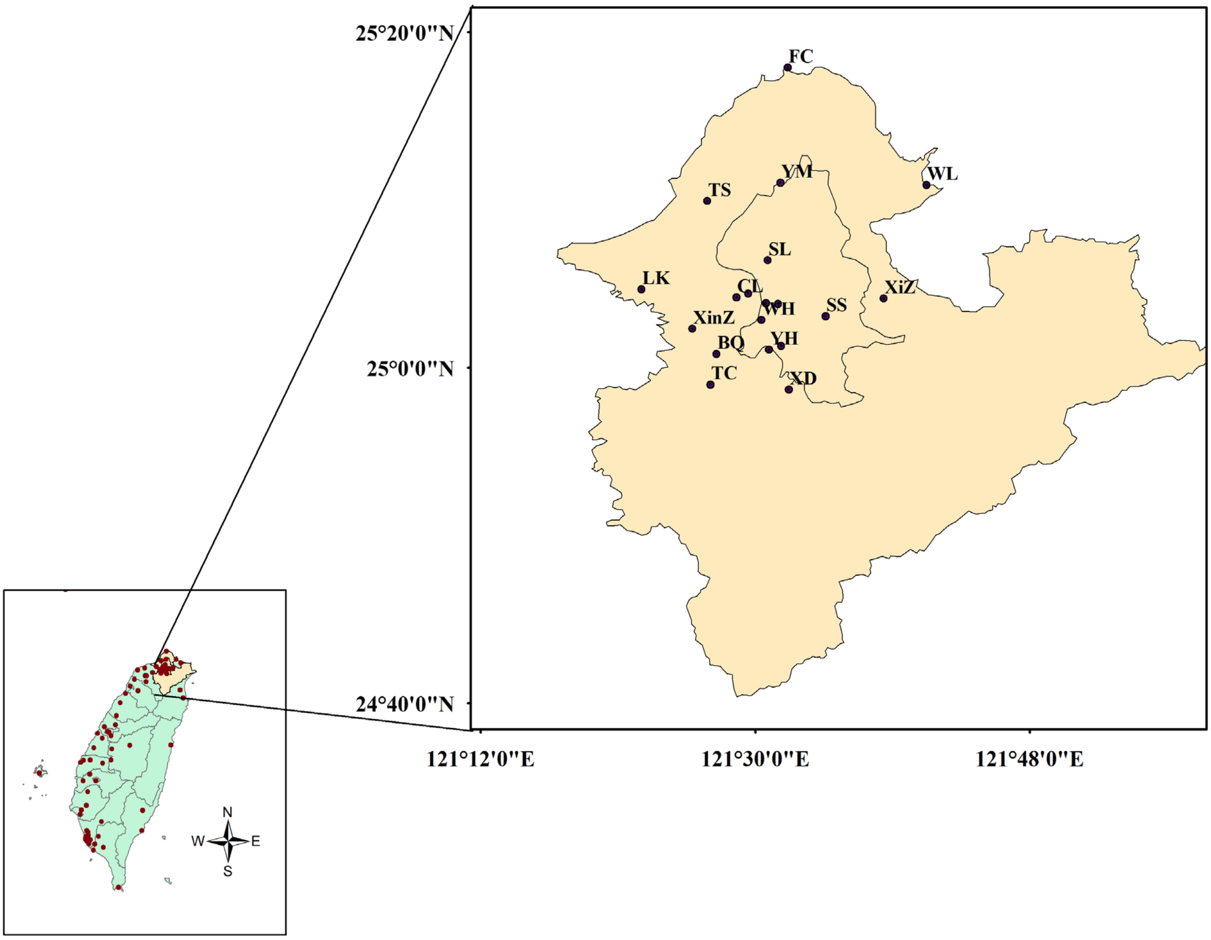

Fig. 1. The location of AQMSs in Taipei and New Taipei Cities (SC (San-chong); TC (Tu-cheng); SL (Shi-lin); DT (Da-tong); ZS (Zhong-shan); GT (Gu-ting); YH (Yong-he); XiZ (Xi-zhi); SS (Song-shan); BQ (Ban-qiao); LK (Lin-kou); TS (Tam-sui); CL (Cai-liao); XD (Xin-dian); XinZ (Xin-zhuang); WH (Wan-hua); WL (Wanli); YM (Yaming Nantional Park); FC (FugueiCape)).

13 general stations (except WL), and 3 traffic stations in Big Taipei City along with the meteorological observations and site specifications were included for analysis.

Briefly, the FRM calibration procedure has been implemented by the contractors of Taiwan EPA across Taiwan since 2014. Due to limited resources, filter-based FRM measurements at only 5 AQMSs in big Taipei were conducted once every three days. In the analysis, 16 AQMSs shared five FRM measurements of $PM_{2.5}$ simultaneously. As a result, 16 resultant calibration equations by the station were generated for every year (Tables S1–S4).

The regression model for the calibration of FRM-like $PM_{2.5}$ measurements from 2014 to 2017 in Big Taipei City is shown in the supplementary data. The measurement data, including ambient temperature, relative humidity, wind speed, rainfall, CO, NO, NO_2, NO_x, O_3, SO_2, CH_4, Non-methane hydrocarbon (NMHC), and total hydrocarbon (THC), were obtained from the Taiwan Air Quality Monitoring Network database for 16 stations. Station profiles such as the station types (ambient/traffic), the height of sampling ports (3.5–13.5 m/17.5 m/19.5–21.5 m), the distance to the nearest main road (1–5.6 m/10–15 m/20–100 m), the types of automatic sampling instruments (VEREWA F701/MetOne 1020) were also collected. Moreover, we incorporated the concurrent atmospheric pressure measurements obtained from the weather bureau stations selected based on the distance to the nearest AQMS. As a lot of meteorological information from AQMSs was missing, all the missing data were imputed by the concurrent measurements from the nearest weather bureau station.

2.2 Statistical Analysis

The SPSS software was used to evaluate temporal (within-site) and spatial (between-site) variability in $PM_{2.5}$ concentrations from FEM and FRM-like measurements at 16 stations. The temporal variation of $PM_{2.5}$ is defined as the variability in $PM_{2.5}$ over time at each AQMS, whereas the spatial variation is defined as the variability in $PM_{2.5}$ across sites at any time. Both FEM and FRM-like $PM_{2.5}$ concentrations had skewed distribution and were normalized by logarithmic transformation. The measurements at daily and yearly time resolutions were evaluated to explore the trends in spatial and temporal variations at short-term and long-term scales.

Pearson's correlation coefficient (r) was used to evaluate the correlations between the variables. Among the variable pairs having $r > 0.7$, only the determinants considered more logically related to $PM_{2.5}$ and/or more consistently and reliably available throughout the duration of the study across selected AQMSs were retained for modeling. The mixed-effects model based on a restricted maximum likelihood estimation procedure was used to examine the relationship between each variable and the log-transformed daily and annual $PM_{2.5}$. The determinants were treated as fixed effects in the model. The AQMSs were treated as random effects to account for potential correlation within repeated measurements at the same AQMS. For $PM_{2.5}$, the mixed-effects model was specified as (Peretz *et al.*, 2002):

$$Y_{ij} = \beta_0 + \beta_1 X_{ij1} + \ldots + \beta_p X_{ijp} + b_1 z_1 + \ldots. + b_k z_k + \varepsilon_{ij} \tag{1}$$

For $i = 1, \ldots, k$ (AQMSs) and $j = 1, \ldots, n_i$ (the repeated day or year of the i^{th} AQMS), where Y_{ij} is the log-transformed $PM_{2.5}$ concentration; β_0 is an overall intercept for the study area that corresponds to mean background $PM_{2.5}$ (log-transformed) when all factors equal zero; $\beta_1, \ldots, \beta_p$ are fixed effects; $X_{ij1}, \ldots, X_{ijp}$ are values of the variables for the i^{th} AQMS on the j^{th} day or year; $b_1, \ldots, b_k$ are AQMSs' random effects; b_i is the i^{th} AQMS random effect, which corresponds to the discrepancy between its intercept and the group intercept β_0; *and* $z_1, \ldots, z_k$ are AQMSs' indicators. ε_{ij} is the residual error associated with i^{th} AQMS on the j^{th} day or year. b_i and ε_{ij} were assumed to be independent and normally distributed, with a mean of 0, and the variances of σ^2_B (between-site) and σ^2_W (within-site), respectively.

Exponential beta [$\exp(\beta)$] value of determinants strengthens the correlation of variables with $PM_{2.5}$ in the model. The statistical significance was set at $p < 0.05$ based on a two-tailed analysis. We created univariate and multivariate models to evaluate the selective variables statistically affecting $PM_{2.5}$. The fitness of each mixed-effects model with selected variables was estimated by the Akaike information criterion (AIC). A lower AIC indicates a better model fit.

3 RESULTS AND DISCUSSION

3.1 $PM_{2.5}$ Concentrations from the Calibrated and Uncalibrated Measurements

Table 1 shows the descriptive statistics of $PM_{2.5}$ concentrations from the FRM-like (calibrated) and FEM (uncalibrated) measurements in the Big Taipei City at 16 AQMSs during 2014–2017. The annual mean concentrations of FRM-like $PM_{2.5}$ were 21.5, 18.5, 17.5, and 16.6 µg m^{-3} in 2014, 2015, 2016, and 2017, respectively, with all the concentrations exceeding the annual standard (15.0 µg m^{-3}) set by Taiwan EPA. In general, the mean $PM_{2.5}$ concentrations from the FEM measurements were approximately 30% significantly ($p < 0.001$) higher than that from the FRM-like measurements at all the stations during 2014–2017. The ZS and LK stations had the highest mean $PM_{2.5}$ concentrations from the FEM and FRM-like measurements, respectively, throughout the duration of the study. The traffic stations, including SC, DT, and YH, did not show higher $PM_{2.5}$ concentrations compared with the concentrations observed at the ambient stations. As shown in Fig. S1, the annual mean concentrations of $PM_{2.5}$ from both the FEM and FRM-like measurements significantly decreased by approximately 22% from 2014 to 2017 at all the AQMSs. The improvement in the air quality in terms of $PM_{2.5}$ is likely attributed to several effective policies implemented by the Taiwan EPA and Environmental Protection Bureau of Taipei. Few of the policies implemented include eliminating two-stroke motorcycles and old-generation diesel vehicles, promoting electric buses and electric motorbikes, emission control measures for the catering industry, stricter standards for boiler emissions, and implementing low-sulfur jet fuel. As shown in Fig. S1, the $PM_{2.5}$ concentrations at CL (31.9 µg m^{-3} in 2014 to 20.5 µg m^{-3} in 2017) from FEM measurements

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Table 1. The descriptive statistics of $PM_{2.5}$ concentrations (µg m^{-3}) with calibrated and un-calibrated measures in Big Taipei city among 16 AQMS for 2014–2017.

Station	Station Type	Measure	n (day)	Mean	SD	GM	25th	75th	Min	Max
San-chong (SC)	Traffic	FRM-like	1,456	18.6	10.5	15.8	11.1	23.9	0.92	83.5
		FEM	1,456	30.1	11.3	28.3	22.2	36.2	10.6	99.0
Tu-cheng (TC)	Ambient	FRM-like	1,450	20.0	11.3	16.9	11.8	25.3	1.25	78.3
		FEM	1,450	24.7	12.7	21.8	15.9	30.4	3.29	88.6
Shi-lin (SL)	Ambient	FRM-like	1,457	17.2	9.39	15.0	10.5	21.5	2.33	78.4
		FEM	1,457	23.2	10.6	21.2	15.7	27.9	6.00	92.4
Da-tong (DT)	Traffic	FRM-like	1,458	16.7	9.48	14.3	9.95	21.3	2.33	67.2
		FEM	1,458	26.8	11.1	24.8	18.8	32.2	9.38	94.7
Zhong-shan (ZS)	Ambient	FRM-like	1,453	18.4	9.76	16.1	11.5	23.0	1.33	88.6
		FEM	1,453	30.9	11.1	29.1	22.7	36.1	12.3	105
Gu-ting (GT)	Ambient	FRM-like	1,445	17.6	10.1	15.0	10.8	22.2	1.43	79.7
		FEM	1,445	20.4	10.9	17.7	12.6	25.6	1.74	85.5
Yong-he (YH)	Traffic	FRM-like	1,452	18.2	9.90	15.7	11.1	23.2	1.79	76.8
		FEM	1,452	22.2	10.9	19.7	14.3	27.6	2.88	89.8
Xi-zhi (XiZ)	Ambient	FRM-like	1,450	18.8	9.75	16.2	12.0	24.0	1.00	77.0
		FEM	1,450	21.6	11.1	18.7	13.9	27.3	0.3	85.2
Song-shan (SS)	Ambient	FRM-like	1,451	17.9	9.37	15.7	10.9	22.7	3.00	73.2
		FEM	1,451	24.1	10.2	22.1	16.3	29.3	7.29	85.0
Ban-qiao (BQ)	Ambient	FRM-like	1,434	19.4	11.2	16.4	11.4	24.8	1.37	77.6
		FEM	1,434	22.7	13.0	19.2	13.5	29.1	0.79	89.1
Lin-kou (LK)	Ambient	FRM-like	1,453	21.8	11.2	19.1	13.9	27.7	1.67	86.5
		FEM	1,453	24.7	11.9	22.1	16.4	30.8	3.21	90.2
Tam-sui (TS)	Ambient	FRM-like	1,450	17.0	9.67	14.4	10.1	21.3	0.50	75.4
		FEM	1,450	22.1	10.9	19.6	14.6	27.2	2.38	89.3
Cai-liao (CL)	Ambient	FRM-like	1,457	18.6	11.0	15.4	10.6	24.9	0.17	75.0
		FEM	1,457	24.8	12.9	21.6	15.5	32.0	2.25	87.6
Xin-dian (XD)	Ambient	FRM-like	1,457	18.3	9.47	16.0	11.6	23.1	2.13	72.3
		FEM	1,457	17.9	10.4	15.0	10.6	23.0	0.83	79.8
Xin-zhuang (XinZ)	Ambient	FRM-like	1,440	19.3	11.7	15.9	10.7	25.4	1.31	76.9
		FEM	1,440	25.0	13.1	21.4	15.6	32.0	2.69	85.3
Wan-hua (WH)	Ambient	FRM-like	1,453	18.7	10.0	16.4	11.4	23.6	2.88	80.4
		FEM	1,453	23.4	10.2	21.5	16.0	28.3	6.63	86.8
Overall	-	FRM-like	23,216	18.5	10.3	15.9	11.2	23.6	0.17	88.6
		FEM	23,216	24.0	11.9	21.2	15.7	30.1	0.30	105

and at XinZ (23.3 µg m^{-3} in 2014 to 14.0 µg m^{-3} in 2017) from FRM-like measurements drastically reduced by 35–40% owing to the operation of Taipei mass rapid transit (MRT) for the Xinzhuang reduced by 35–40% owing to the operation of Taipei mass rapid transit (MRT) for the Xinzhuang line in 2014. In addition, the lowest decline in $PM_{2.5}$ concentrations was observed at LK (12.5%) and XD (7%) from the FEM and FRM-like measurements, respectively. The FEM and FRM-like measurements presenting such a dissimilarity in the reduction of $PM_{2.5}$ levels measured at the stations may mask information on the effectiveness of region-specific ambient air quality management.

3.2 Variations in $PM_{2.5}$ Concentrations from the FEM and FRM-like Measurements

Table 2 shows σ_B^2, σ_W^2, and total variance (σ_{Total}^2) of daily and annual FEM and FRM-like $PM_{2.5}$ concentrations at 16 AQMSs during 2014–2017. The daily FEM $PM_{2.5}$ concentrations were dominated by the within-site variability (> 90% of σ_{Total}^2) rather than the between-site variability. When the daily FRM-like data was analyzed in the null model (all factors of fixed effects equal zero), the between-site variability (σ_B^2 = 0.004; 1.29% of σ_{Total}^2) in $PM_{2.5}$ concentrations decreased

Table 2. The between- (σ_B^2) and within-site (σ_W^2) variability and total variance (σ_{Total}^2) in $PM_{2.5}$ concentrations for FEM and FRM-like measures in daily and yearly time scales among 16 AQMS for 2014–2017.

Time resolution	Measure	n	σ_W^2	σ_B^2	σ_{Total}^2	σ_W^2 (%)	σ_B^2 (%)
Daily	FEM	23,214	0.245	0.027	0.272	90.2	9.84
	FRM-like		0.339	0.004	0.343	98.7	1.29
Yearly	FEM	64	0.016	0.014	0.030	52.3	47.8
	FRM-like		0.020	0	0.019	100	0

by ~8% of total variance compared with FEM data (σ_B^2 = 0.027; 9.84% of σ_{Total}^2) as shown in Table 2. The increase of σ_W^2 (=0.339; 98.7% of σ_{Total}^2) in FRM-like $PM_{2.5}$ could be also observed compared with that in FEM $PM_{2.5}$ (σ_W^2 = 0.245; 90.2% of σ_{Total}^2).

While fitting such linear regression models, the calibration procedure limits the inherent variation in $PM_{2.5}$ concentrations obtained from the AQMSs at various locations, which may reduce spatial variation in $PM_{2.5}$ concentrations. Basically, the FRM measurements of $PM_{2.5}$ at five AQMSs (SL, WH, XZ, BQ, and Taoyuan) were used to calibrate single or multiple FEM measurements at the corresponding AQMSs through linear regression models and coefficient of determination (R^2, mean = 0.92, range = 0.83–0.97) during 2014–2017 (Tables S1–S4). For instance, the FRM measurement at one station (WH) was shared by the FEM measurements at a maximum of 8 AQMSs for calibration, where R^2 ranged from 0.86 to 0.97 in 2014 (Table S1). When we compared daily variations in $PM_{2.5}$ concentrations from the FRM and FEM measurements during 2014–2017, σ_{Total}^2 and coefficient of variation (CV) and σ_W^2 of $PM_{2.5}$ concentrations from the FRM measurements were larger than those from the FEM measurements (Table S5). Hence, it can be stated that the calibration approach of FRM-like $PM_{2.5}$ measurements directly decreases the spatial heterogeneity in $PM_{2.5}$ concentrations across sites.

For annual $PM_{2.5}$, the within-site (σ_W^2 = 0.016) and between-site (σ_B^2 = 0.014) variance in the FEM measurements were similar. However, the between-site variance in the FRM-like measurements became negligible (σ_B^2 = 0%) after the calibration process. The within-site variation (σ_W^2 = 0.245, accounting for 90.2% of σ_{Total}^2) in daily $PM_{2.5}$ concentrations was much higher than that (σ_W^2 = 0.016, accounting for 52.3% of σ_{Total}^2) in annual $PM_{2.5}$ concentrations, indicating the importance of temporal variation in $PM_{2.5}$ concentrations for studying short-term $PM_{2.5}$ exposure. Che *et al.* (2015) indicated that the within-group exposure variability is larger than the between-group variability for the daily exposure of children to $PM_{2.5}$. Our recent study also reported that the within-subject variability (81.3% of σ_{Total}^2) in personal exposure concentrations of $PM_{2.5}$ was higher than the between-subject variability in concentrations (18.7% of σ_{Total}^2) for university students (Hsu *et al.*, 2020). In contrast, long-term (annual) concentrations of $PM_{2.5}$ were fairly attributable to the between-site or between-group variability (47.8%). As a result, some advanced exposure models with the spatial variables have been used to estimate $PM_{2.5}$ exposure. For instance, the land-use regression model incorporating annual $PM_{2.5}$ levels from the monitoring stations and geographic information has been successfully developed to predict ambient $PM_{2.5}$ concentrations for exposure estimates in epidemiological studies (Eeftens *et al.*, 2012; Wu *et al.*, 2017). However, $PM_{2.5}$ concentrations from FEM measurements are more reliable to be incorporated in geographic-based prediction models, while the annual FRM-like $PM_{2.5}$ data has almost zero between-site variability (i.e., σ_B^2 = 0). To minimize the measurement error, the calibration of FRM-like $PM_{2.5}$ is still suggested after the exposure estimates in the health risk analysis have been done.

3.3 Factors Determining Variability in $PM_{2.5}$ Concentrations

Table 3 shows the definition of variables for 16 AQMSs. The information about these variables was obtained from the Taiwan EPA site and the nearest weather bureau. FEM $PM_{2.5}$ concentrations and 12 variables associated with meteorology, gaseous air pollutants, and sampling site characteristics were included in the models. Since FRM-like $PM_{2.5}$ concentrations presented an extremely low σ_B^2, this data was not evaluated in the mixed-effects model.

Table 4 shows the model coefficients for the selected variables correlated with daily FEM $PM_{2.5}$ determined by the univariate and multivariate analysis. By performing the univariate analysis in the model, we found that the $PM_{2.5}$ concentrations decreased significantly from 2014 to 2017,

Table 3. Definition of variables for 16 AQMS obtained from Taiwan EPA site and Weather Bureau.

Variable (unit)	Type of variable	Number of measurements	Description
Year	Categorical	23,214	2014 2015 2016 2017
Meteorological factors			
Ambient temperature (°C)	Continuous	23,192	Range: 3.64–37.0 Mean: 23.9
Ambient Pressure (Pa)	Continuous	19,228	Range: 962–1034 Mean: 1010
Relative humidity (%)	Continuous	23,193	Range: 32.5–100 Mean: 73.9
Wind speed (m s^{-1})	Continuous	23,181	Range: 0.1–14.7 Mean: 1.6
Rainfall (mm)	Dichotomous	23,156	Range: 0.0–379 Mean: 0.6
Air pollutant factors			
NO_2 (ppb)	Continuous	23,202	Range: 0.22–67.5 Mean: 20.4
O_3 (ppb)	Continuous	20,270	Range: 1.22–85.9 Mean: 27.3
SO_2(ppb)	Continuous	23,121	Range: 0.22–22.6 Mean: 3.1
Sampling site factors			
Station type	Categorical	23,214	0: Traffic station 1: General station
Height of sampling port (m)	Ordinal	23,214	1: 3.5–13.5 2: 17.5 3: 19.5–21.5
Distance to main road (m)	Ordinal	21,757	1: 1–5.6 2: 10–15 3: 20–100
Type of instrument	Categorical	23,214	0: VEREWA F701 1: MetOne 1020

when compared with the reference concentration in 2017. The meteorological factors including ambient temperature, relative humidity, wind speed, and rainfall were negatively correlated ($p <$ 0.001) with ambient $PM_{2.5}$, which explained 18.3% [(0.246–0.201)/0.246] of the within-site variability and 6.63% [(0.027–0.025)/0.027] of the between-site variability (Table S6). The ambient pressure was not included in the model for daily $PM_{2.5}$ estimates because of its high correlation with ambient temperature. The factors such as gaseous air pollutants (NO_2, O_3, and SO_2) were positively correlated ($p < 0.001$) with ambient $PM_{2.5}$, explaining 45.8% [(0.246–0.133)/0.246] of the within-site variability and 26.8% [(0.027–0.020)/0.027] of the between-site variability (Table S6). Among sampling site factors, the only variable significantly affecting $PM_{2.5}$ concentrations was the type of instrument used for measurement, explaining 0% of the within-site variability but 19.5% [(0.027–0.021)/0.027] of the between-site variability (Table S6). It was observed that the VEREWA F701 sampler presented a higher value of 17% than the MetOne 1020 sampler. By performing the multivariate analysis in the model, we found that all the factors significantly affected concentration variability, explaining 50.2% [(0.245–0.122)/0.245] of the within-site variability and 15.3% [(0.027–0.023)/0.027] of the between-site variability (Table S6). The AIC scores from each mixed-effects model were shown in Fig. 2. We found that the selective model considering all the significant variables can predict 55% of ambient $PM_{2.5}$ concentrations. Among all the variables, NO_2, O_3, and SO_2 were the predominant variables, explaining 22.1%, 13.5%, and 16.1% of σ_{Total}^2 of $PM_{2.5}$, respectively.

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Table 4. Model coefficients for meteorological, air pollutant and sampling site factors associated with daily ambient $PM_{2.5}$ with FEM measurements.

Variables	univariate		multivariate	
	β	Exp(β) (S.E.)	β	Exp(β) (S.E.)
Year				
2014	0.274	1.32 (1.01)**	0.171	1.19 (1.01)**
2015	0.147	1.16 (1.01)**	0.109	1.12 (1.01)**
2016	0.036	1.04 (1.01)**	0.064	1.07 (1.01)**
2017	-	-	-	-
Meteorological factors				
Ambient temperature	–0.016	0.984 (1.00)**	-0.008	0.992 (1.00)**
Relative humidity	–0.010	0.990 (1.00)**	-0.008	0.992 (1.00)**
Wind speed	–0.162	0.850 (1.00)**	-0.051	0.948 (1.00)**
Rainfall	–0.008	0.992 (1.00)**	-0.003	0.997 (1.00)**
Air pollutant factors				
NO_2	0.038	1.038 (1.00)**	0.035	1.036 (1.00)**
O_3	0.014	1.014 (1.00)**	0.017	1.018 (1.00)**
SO_2	0.159	1.173 (1.00)**	0.090	1.091 (1.00)**
Sampling site factors				
Station type				
Traffic	0.150	1.162 (1.106)	-	-
Ambient	-	-	-	-
Height of sampling port				
3.5–13.5 m	0.014	1.014 (1.105)	-	-
17.5 m	–0.089	0.915 (1.113)	-	-
19.5–21.5 m	-	-	-	-
Distance to main road				
1–5.6 m	0.080	1.084 (1.097)	-	-
10–15 m	0.065	1.067 (1.097)	-	-
20–100 m	-	-	-	-
Type of instrument				
VEREWA F701	0.159	1.173 (1.077)*	0.035	1.04 (1.09)
MetOne 1020	-		-	-

The meteorological factors have a strong influence on the pollutants, leading to daily fluctuations in the pollutant levels. These factors have been shown to explain 50–60% of σ_{Total}^2 of winter concentrations in China in a previous study by Zhang *et al.* (2015). The elevated $PM_{2.5}$ concentrations with lower temperature and higher atmospheric pressure are usually observed because the lower mixing layer and higher frequency of thermal inversion in winter can restrict atmospheric dispersion and thereby trap organic and inorganic matter in the particles. It was observed that the decrease in wind speed due to unfavorable atmospheric diffusion could lead to an increase in $PM_{2.5}$ concentrations (Liao *et al.*, 2018). The long-range transport usually observed in Taipei during the cold season may be, in part, attributable to the elevated $PM_{2.5}$ concentrations (Kuo *et al.*, 2013; Hsu *et al.*, 2019). The positive correlation of $PM_{2.5}$ with gaseous air pollutants in our study confirmed the enhanced levels of $PM_{2.5}$ due to the chemical reactions in the atmosphere involving precursor gases of NO_2 and SO_2. O_3 was significantly and positively correlated with $PM_{2.5}$, which probably could be explained by the covariance of VOCs. However, the precursor formation and meteorological conditions can add complexity to the relationship between both the air pollutants. Ito *et al.* (2007) have reported that the correlation between O_3 and $PM_{2.5}$ changed with the season (positive in summer and negative in winter). In a previous study by Liu *et al.* (2013), it was highlighted that the use of different aerosol samplers (MetOne 1020 vs VEREWA-F701) for measuring $PM_{2.5}$ concentrations might result in a potential bias of exposure variability. It was not surprising that the height of the sample port, distance to the nearest main road, and station type were not significantly associated with ambient $PM_{2.5}$ because the inherent confounders were not available for adjustment. Quang *et al.* (2012) indicated that

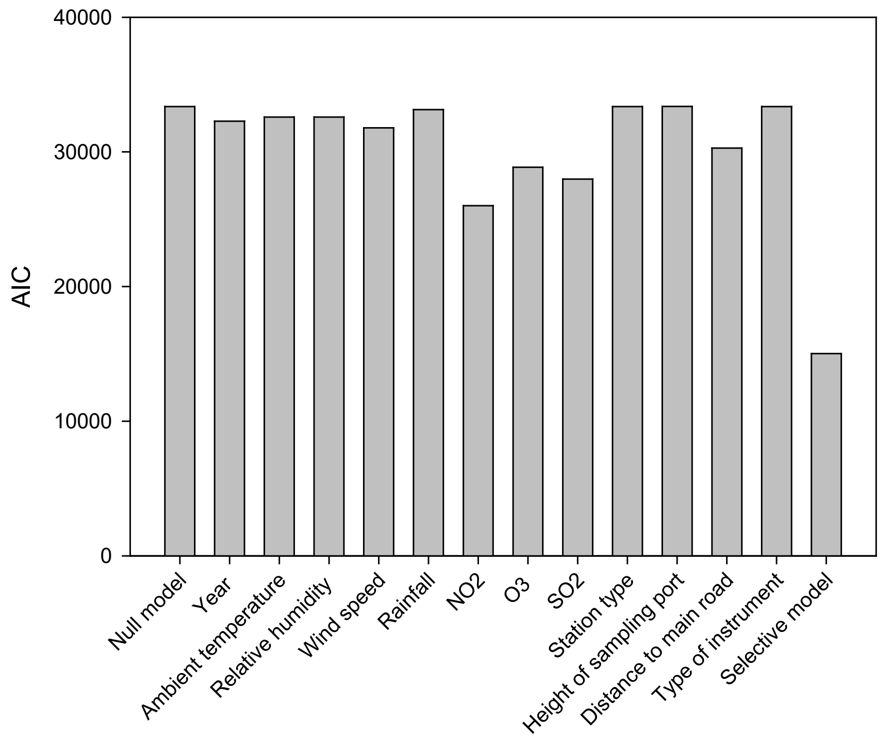

Fig. 2. AIC scores for the selected variables associated $PM_{2.5}$ concentrations using mixed-effect models.

the $PM_{2.5}$ concentrations decreased with the increase in the height of the office building; however, the vehicle emissions, particle formation, flow patterns around the building envelope, and street profile were the important determinants that should be taken into account for comparison. In addition, the differences in the aerosol instruments (MetOne 1020 and VEREWA F701) used in our study and the variations in the sampling factors (i.e., height of the sample port, distance to the nearest main road, and station type) led to variations in $PM_{2.5}$ concentrations. For annual $PM_{2.5}$, SO_2 was significantly correlated with $PM_{2.5}$ in addition to ambient temperature and time (in years) (Table S7). In the population-based study associated with exposure to $PM_{2.5}$, meteorological conditions (i.e., temperature) are often adjusted in the model (Luo *et al.*, 2016). Our results showed SO_2 as an important factor showing a significant positive correlation with both daily and annual $PM_{2.5}$. We suggest that SO_2 should be taken into account in the future for health risk analysis associated with $PM_{2.5}$ exposure for adjustment.

4 CONCLUSIONS

In this study, we highlighted that the FRM-like $PM_{2.5}$ measurements with negligible between-site variability might lead to non-differential misclassification of exposure. The gaseous pollutants (such as NO_2, O_3, and SO_2) are significant factors affecting the spatial and temporal variations in ambient $PM_{2.5}$ concentrations, which should be incorporated in the health risk assessment model. We indicated that the FEM measurements of $PM_{2.5}$, rather than the FRM-like measurements, at the AQMSs are mainly applicable for exposure estimates in epidemiological studies. Then, the resultant exposure estimates of $PM_{2.5}$ are further calibrated with the FRM measurements to minimize the measurement errors.

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DISCLAIMER

The authors declare no conflict of interest.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at https://doi.org/10.4209/aaqr.2020.05.0217

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