

# An Integrated Approach to Characterize Temporal–Spatial Variations in PM<sub>2.5</sub> Concentrations at the Ground Level and Its Implication on Health Impact Assessments

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Ho M-S, Lin M-Y, Wang J-D, Young L-H, Hsu H-T, Hwang B-F and Tsai P-J (2022) An Integrated Approach to Characterize Temporal–Spatial Variations in PM<sub>2.5</sub> Concentrations at the Ground Level and Its Implication on Health Impact Assessments. Front. Environ. Sci. 10:901754. doi: 10.3389/fenvs.2022.901754 Although PM<sub>2.5</sub> concentrations measured by the governmental air quality monitoring station (AQMS) have been widely used for conducting exposure assessments, it might be not able to reflect the residents' exposures, especially for those associated with ground emissions. The present study was conducted in a city area for 1 year. A mobile monitoring station (MMS) was established to measure the  $PM_{2.5}$  concentrations at the ground level. A significant linear relationship ( $R^2 = 0.53$ ) was found between the MMS-measured concentrations and the corresponding concentrations obtained from the AQMS (15 m above the ground level), and the former was ~ 1.11 times (95% Cl: 1.08-1.15) in magnitude higher than that of the latter. To characterize the spatial variation of the area, the MMS-measured values were further classified into three different regions. A consistent trend was found in the present study for all collected data as industry region≒urban region > harbor region. The aforementioned results clearly indicate that the residents' ambient PM2.5 exposures do have spatial differences. Seven-year AQMS-measured concentrations (i.e.,  $AQMS_{7-vr}$ ) were used to establish the long-term  $PM_{2.5}$ concentrations at the ground level (i.e.,  $MMS_{7-vt}$ ) of the three different regions using the linear regression equations obtained from the MMS and AQMS. Health impact functions and local health data were used to quantify the PM2.5-attributable health burden for both AQMS7-vr and  $MMS_{7-yr}$ , respectively. Results show that the former is ~ 10.4% lower in magnitude than the latter in the estimated lung cancer death attributed fraction (AF). In particular, the decrease of unit PM<sub>2.5</sub> (µg/m<sup>3</sup>) would lead to a 0.75 and 0.71% decrease in the estimated AF of lung cancer death for AQMS<sub>7-vr</sub> and MMS<sub>7-vr</sub>, respectively. As a result, directly using AQMS<sub>7-vr</sub> would lead to an underestimation of ~ 1,000 lung cancer deaths annually in Taiwan in comparison with those using MMS7-vr. The aforementioned results clearly indicate the importance of characterizing ground-level exposures for assessing the health impact of residents, and the methodology developed by the present study would be helpful for solving the aforementioned problem.

Keywords: PM2.5, mobile monitoring station, air quality monitoring station, exposure assessment, health impact assessment

# INTRODUCTION

It has been reported that air pollution is responsible for  $\sim 2.9$ million deaths per year accounting for ~ 35% of all environmental pollution-related deaths (Fuller et al., 2019). The International Agency for Research on Cancer (IARC) has classified outdoor air pollution as Group 1 carcinogen (i.e., carcinogenic to humans) (Straif et al., 2013). Among various particulate matters (PMs), many pieces of evidence have shown that PM<sub>2.5</sub> (i.e., PM with a particle size less than 2.5  $\mu$ m) has a much stronger impact on human health than those with greater particle sizes (Environmental Protection Agency U.S, 2019). PM<sub>2.5</sub> is responsible for many adverse health effects, including lung cancer, chronic obstructive pulmonary disease (COPD), ischemic heart disease (IHD), strokes, and acute lower respiratory infections (ALRIs). (Mustafic et al., 2012; Forouzanfar et al., 2016; Monrad et al., 2017). Therefore, the control of atmospheric PM<sub>2.5</sub> concentrations has been considered as the primary concern in many countries. However, it should be noted that the initiation of appropriate abatement strategies for PM2.5 should start with knowing the correct PM2.5 exposure levels and health impact on the residents in the target area. Considering the existence of the intrinsic spatial and temporal variations in residents'  $PM_{2.5}$ exposure concentrations, developing suitable exposure assessment (EA) methodologies has become an important issue in the environmental health science field.

At present, the methodologies used for conducting PM<sub>2.5</sub> EA can be classified into two categories: the non-ground level (also called macroenvironment) and ground level (microenvironment) (Straif et al., 2013; Dias and Tchepel, 2018; Li et al., 2018; Caplin et al., 2019; Environmental Protection Agency U.S, 2019). For non-ground-level EA methods, exposure data can be obtained from governmental air quality monitoring stations (AQMS) (i.e., fixed monitoring stations), model estimation, satellite remote sensing, etc. (Ozkaynak et al., 2013; Environmental Protection Agency U.S, 2019). The aforementioned methodologies are widely used in conducting EA on a large scale (such as on the whole country level, or even acrosscountry level) and assuming the uniformity of PM2.5 concentrations in each divided subarea. For example, PM<sub>2.5</sub> AQMS data from many countries were collected by the World Health Organization (WHO) for assessing the global exposure and health burden assuming the uniformity of PM2.5 concentrations at a city level (W.H.O., 2012). However, the aforementioned methods might not be feasible for conducting PM<sub>2.5</sub> EA in a small area (such as at a city level) because the spatial heterogeneity might have a significant impact on the residents' PM<sub>2.5</sub> exposure concentrations (Fishbain et al., 2017; Malings et al., 2020).

It is known that urban traffic accounts for >50% of the total  $PM_{10}$  emissions. In comparison with  $PM_{10}$ , urban traffic has an even greater contribution to  $PM_{2.5}$  concentrations at the ground level (Han and Naeher, 2006). The main traffic emission sources include road traffic, airport, and port operations (Kingham et al., 2013; Mazaheri et al., 2018). Among them, urban ground  $PM_{2.5}$ 

concentrations are mainly influenced by road traffic contributed by both traffic exhaust and non-traffic exhaust (e.g., brake wear and road dust) (Jeong et al., 2019). Considering the proximity of residents to ground PM<sub>2.5</sub> emission sources (such as traffic), higher exposure concentrations at the ground level are expected than those obtained from the non-ground level (e.g., AQMS). Therefore, many methodologies have been developed to measure ground-level PM2.5 concentrations (such as the mobile monitoring station (MMS) (Hankey and Marshall, 2015; Okokon et al., 2017; Lin et al., 2018; Cheng et al., 2019; Shen and Gao, 2019), air box (Lee et al., 2019; Lin et al., 2020), and modeling estimation (Hu et al., 2016; Jung et al., 2018)) for better characterizing residents' exposures in the small area of interest. Although the air box and modeling methods are less expensive and provide real-time information, many limitations are involved if they are used for conducting EA, including the accuracy and reliability of the collected data (Cabada et al., 2004; Snyder et al., 2013; Castell et al., 2017; Morawska et al., 2018; Williams et al., 2019; Li et al., 2020; Zamora et al., 2020; Environmental Protection Agency U.S, 2021). For the MMS, it has been found to be more accurate and reliable than the aforementioned two methodologies. On the condition of the existence of an AQMS in the target area, the use of the MMS method provides the possibility for better characterizing spatiotemporal personal exposures in а given microenvironment by combining with the AQMS data (Hankey and Marshall, 2015; Droge et al., 2018; Piotrowicz and Polednik, 2019).

In Taiwan, at least one AQMS is installed in every city or township for characterizing PM2.5 concentrations in different areas. Although concentrations obtained from the AQMS provide longitudinal information of the area, however, they are inadequate to characterize the residents' PM2.5 exposures in different regions of the area because of their intrinsic spatial heterogeneity (Borge et al., 2016). Therefore, developing new monitoring strategies with the aid of long-term AQMS data to characterize the residents' both temporal and spatial exposures has become an important issue for conducting PM<sub>2.5</sub> EAs. In principle, the development of predicting models between PM2.5 concentrations of the AQMS and MMS would be helpful for establishing the residents' long-term PM2.5 exposure concentrations at the ground level in different regions. However, it should be noted that before the establishment of the aforementioned predicting models, possible interference factors should be considered, including meteorological factors (e.g., temperature, humidity, (RH), wind speed, etc.) (Ye et al., 2018; Zhang et al., 2018; Lee et al., 2020; Rittner et al., 2020; Yousefian et al., 2020; Yang et al., 2022) and other co-emission pollutants (e.g., primary pollutants of NO2, SO2 etc. and secondary pollutants of O<sub>3</sub>) (Saraswat et al., 2013; Zhang et al., 2018; Guo et al., 2019).

Since  $PM_{2.5}$  concentration plays a significant role in human health, conducting a health impact assessment (HIA) would provide better evidence to help decision-makers for developing proper control strategies (W.H.O., 2022). For any given air pollutant, the calculation of the burden of disease (BOD) is widely adopted for conducting the HIA (van der Kamp and



Bachmann, 2015; W.H.O., 2016; Maji et al., 2017; Malmqvist et al., 2018; Wu et al., 2018). In principle, four components should be collected for conducting the HIA: the exposure and reference concentrations of the air pollutant of interest, the resultant health outcomes, associated concentration–response functions (CRFs) and attributed fractions (AFs), and incidence rate (IR) of the health effect. CRFs can be determined using the novel Global Exposure Mortality Model (GEMM). If the exposure and reference concentrations of the air pollutant of interest were measured, the CRF can be determined, and finally, the burden of disease associated with the given air pollutant can be estimated (Ostro and WHO, 2004). This process has been widely adopted by WHO and others for conducting HIAs associated with PM<sub>2.5</sub> exposures.

The present study is set out to establish an effective methodology for assessing  $PM_{2.5}$  exposures at the ground level in an urban region. Considering the existence of an AQMS, an MMS was established to collect concentrations of  $PM_{2.5}$  at the ground level of different regions of the study area to characterize its spatial variation. Since continuous longitudinal data were collected by the AQMS, the AQMS data were used to determine the temporal variation in  $PM_{2.5}$  concentrations in the study area. By combining the obtained MMS data with AQMS results, both temporal and spatial variations in  $PM_{2.5}$  concentrations were characterized. Finally, HIAs were conducted to quantify the  $PM_{2.5}$ -attributable health burden for those data simply considering the temporal variation (i.e., directly obtained from the AQMS) and those considering both temporal and spatial variations (i.e., by

combining both those directly obtained from the AQMS and measured by the MMS) of different regions of the study area. The obtained  $PM_{2.5}$ -attributable health burdens were used for showing the importance of characterizing temporal and spatial variations in conducting  $PM_{2.5}$  exposure and HIAs.

#### MATERIALS AND METHODS

#### Study Area

The whole study was conducted in the Shalu area (including Shalu, Wugi, and Longjing districts with a total area of  $95 \text{ km}^2$ ), located in west Taichung City (the second largest city in Taiwan), with a population of 220,000 residents, 216,000 vehicles (locomotives accounting for ~ 58%), and five industrial/science parks (~ 1,300 manufacturing industries) in 2014. From 2014–2019, there were 5.1, 5.7, and 13.7% increases annually for the number of residents, vehicles, and manufacturing industries, respectively. The region is adjacent to three main traffic roads including one highway (Chen et al., 2015), comprising one coal-fired power plant, one big steel factory, and one harbor located on the west (Kuo et al., 2014) (**Figure 1**).

#### PM<sub>2.5</sub> Monitoring Stations

On the north-western side of the Shalu area, one AQMS (latitude: 24.22563, longitude: 120.5688.) operated by the Taiwan EPA has been installed. In the present study, an MMS was established to collect concentrations of  $PM_{2.5}$  at the ground level of different regions of the study area.

Station	AQMS	MMS		
Station height	15 m above the ground level	Ground level		
Monitoring instrument	BAM-1020 (beta-rays)	DUSTTRAK II Aerosol Monitor 8,530 (90° light scattering)		
Monitoring frequency	per hour	real-time (1s-1 h)		
Monitoring resolution	2 µg/m <sup>3</sup>	1 µg/m <sup>3</sup>		
Calibrated concentrations by reference to FRM/FEM <sup>a</sup> method	$AQMS_c = 0.96*AQMSm - 9.33$ ( $R^2 = 0.92$ ) (Environmental Protection Administration, T.R.O.C, 2020)	MMS <sub>c</sub> = 0.33*MMS <sub>m</sub> + 2.25 (R <sup>2</sup> = 0.859) (Yanosky et al., 2002; TSI_Incorporated, 2013)		

TABLE 1 | Basic information for the AQMS and MMS used in the present study for measuring PM2.5 concentrations.

<sup>a</sup>FRM/FEM, Federal Reference Methods/Federal Equivalent Methods.

#### The Air Quality Monitoring Station

The AQMS is 15 m above the ground level with a sampling inlet at 19.5 m in height. The main monitoring items include fine suspended particulates (PM<sub>2.5</sub>), coarse suspended particulates (PM<sub>10</sub>), carbon monoxide (CO), sulfur dioxide (SO<sub>2</sub>), ozone (O<sub>3</sub>), nitrogen dioxide (NO<sub>2</sub>), and meteorological data (UVB, rainfall, wind direction and wind speed, temperature, and humidity). The installed PM<sub>2.5</sub> monitoring instrument (model BAM-1020, Met One Instruments Inc., Grands Pass, OR) uses beta-rays for detecting the relative concentration of PM<sub>2.5</sub> (detection range: 0–10 mg/m<sup>3</sup>, resolution:  $2 \mu g/m^3$ , beta-ray source: C-14, filter paper: glass fiber, capture efficiency: 0.3  $\mu$ m particulate matter> 99.999%, sampling flow rate: 16.7 LPM) once per hour automatically.

#### The Mobile Monitoring Station

An electric car (LUXGEN MPV EV+, battery module: lithiumion) was chosen as a mobile platform for the MMS to avoid influences from vehicle self-emissions. The MMS is equipped with a DUSTTRAK II Aerosol Monitor (model 8,530, TSI Inc., St. Paul, MN, United States; detection range: 0.001-150 mg/ m<sup>3</sup>, resolution:  $1 \mu g/m^3$ , sampling flow rate: 3 L/min, time resolution: one reading per second) to monitor PM<sub>2.5</sub> realtime concentrations and a GPS to record the latitude-longitude information. To prevent collected samples from being affected by road contaminants, the sampling inlet of the MMS is set at a height of 2.2 m and air samples are collected isokinetically. Table 1 shows the information on the station height, monitoring instrument, data collection frequency, and resolution for both the AQMS and MMS. Considering the intrinsic differences in the monitoring instruments of the AQMS and MMS, both the collected data (i.e., AQMS<sub>m</sub> and MMS<sub>m</sub>) were calibrated (i.e., AQMS<sub>c</sub> and MMS<sub>c</sub>) by reference to the Federal Reference Methods/Federal Equivalent Methods (FRM/FEM) (Environmental Protection Agency U.S, 2022).

#### Sampling Campaign

A sampling route ~ 80 km in length was designed for the MMS to collect  $PM_{2.5}$  concentrations in the study area. Samplings were conducted from September 2013 to August 2014 covering the four seasons. For each sampling day, samplings were conducted approximately from 6:00 to 17:00 and 18:00 to 23:00 during the daytime and nighttime, respectively. Details of the MMS and its QA/QC can be found in our previous work (Lin et al., 2018). To compare with AQMS-collected data (i.e., hourly data), all MMScollected data were further processed as the hourly average values. All collected MMS data in the study area were classified into three regions as follows based on the obtained GPS records: the urban region, industrial region, and harbor region (**Figure 2**). **Table 2** shows the details of the whole MMS sampling campaign. In the present study, the corresponding AQMS data were also recorded in accordance with the aforementioned MMS sampling campaign.

#### **Statistic Methods**

Since the obtained  $PM_{2.5}$  concentrations were lognormally distributed, logarithmic conversion was applied to both AQMS and MMS data. *One-way ANOVA/paired t/t-tests* were applied to examine the differences between the MMS and the AQMS in seasons, day/night, commute periods, and different areas.

To develop predicting models for establishing residents' long-term PM<sub>2.5</sub> exposure concentrations at the ground level in different regions, MMS-measured data were compared with those obtained from the AQMS. Before the establishment of the aforementioned predicting models, possible interference factors were first identified, including three atmospheric variables [temperature (TEMP), relative humidity (RH), and wind speed (WS)], three primary pollutants (CO, NO<sub>x</sub>, and SO<sub>2</sub>), and one secondary pollutant (O<sub>3</sub>) according to literature reviews (Cabada et al., 2004; Borge et al., 2016; Mazaheri et al., 2018; Adams et al., 2020; Liu et al., 2020). The hierarchical regression analysis was used to further confirm interference factors for the final predicting models. All data aggregation and statistics were performed using the Excel 365 software (Microsoft), R software (R x64 3.6.2) (R-Core-Team, 2019), and SPSS17 (IBM SPSS, 2008).

#### **Conducting Health Impact Assessment**

In the present study, ground-level  $PM_{2.5}$  concentrations derived from predicting models were used for deriving the  $PM_{2.5}$ mortality hazard ratio (HR) through the application of the GEMM as follows:

HR = GEMM(z)

$$= exp[\theta * \log(z/\alpha + 1)/(1 + exp[-(z - \mu)/\nu])], \quad (1)$$



TABLE 2 Description of the whole MMS sampling campaign classified into the whole sampling days/hours by the season, day/night, rush/non-rush, and area.

	n	Season <sup>a</sup>		Day/night <sup>b</sup>		Rush_or_not <sup>c</sup>		Region				
		Fall	Winter	Spring	Summer	Daytime	Nighttime	Rush	Non-rush	Urban	Industry	Harbor
Days	59	16	19	8	19	56	49	58	41	59	_	_
Hours	694	148	233	78	235	465	229	584	110	330	248	116

<sup>a</sup>Fall: Sep. 2013—Nov. 2013; winter: Dec. 2013–Feb. 2014; spring: Mar. 2014-May 2014; summer: Jun. 2014-August. 2014.

<sup>b</sup>Daytime: am 06-pm 17; nighttime: pm 18-am 05.

<sup>c</sup>Rush hour: am7-9 and pm18-20; non-rush hour: excluding rush hours.

TABLE 3   Values for the parameter used in the GEMM.									
Cause of death	Age range	Parameters							
		θ	SE $\theta^a$	a	μ	ν			
Lung cancer	>25	0.2942	0.06147	6.2	9.3	29.8			
LRI	>25	0.4468	0.11735	6.4	5.7	8.4			
COPD	>25	0.251	0.06762	6.5	2.5	32			
Stroke	>25	0.272	0.07697	6.2	16.7	23.7			
IHD	>25	0.2969	0.01787	1.9	12	40.2			

<sup>a</sup>SE, standard error of  $\theta$ , which is used for 95%Cl, uncertainty estimation. Parameters quoted from GEMM, study (with Chinese male cohort) (Burnett et al., 2018).

where  $z = \max(0, C - C_{rf}), C =$  the exposed PM<sub>2.5</sub> concentration, and  $C_{rf} =$  the counterfactual PM<sub>2.5</sub> concentration (=2.4  $\mu$ g/m<sup>3</sup>).

 $\theta$ , SE $\theta$ ,  $\alpha$ ,  $\mu$ , and  $\nu$  = parameters can be determined based on the cause of death (for the present study: lung cancer) by referring to **Table 3**.

Since the mortality probability of lung cancer is quite small, the resultant HR could serve as a surrogate for the relative risk (RR) (Burnett et al., 2018). The attributed fraction for the population (AF) of lung cancer can be calculated as follows (Ostro and WHO, 2004):

$$\mathbf{AF} = (\mathbf{RR} - 1)/\mathbf{RR}.$$
 (2)

## **RESULTS AND DISCUSSION**

# Comparisons of AQMS- and MMS-Measured PM<sub>2.5</sub> Concentrations

 $PM_{2.5}$  concentrations of the whole sampling year, seasonal, diurnal, and rush/non-rush hour obtained from both the AQMS and MMS are compared and shown in **Table 4**. From the whole sampling year aspect, MMS results (GM = 25.9  $\mu$ g/m<sup>3</sup>; 95% CI = 24.8–27.1  $\mu$ g/m<sup>3</sup>) are significantly higher than those of

Temporal/spatial	Description	n	MMS	AQMS	MMS/AQMS <sup>a</sup>
			GM (95%	Ratio (95% CI)	
Season					
Fall	Sep. 2013–November. 2013	148	18.5 (17.3-19.8)	22.2 (21.0-23.6)	0.83 (0.79-0.87)
Winter	Dec. 2013–February. 2014	233	33.1 (30.3-36.2)	29.3 (27.1-31.7)	1.13 (1.08-1.19)
Spring	Mar. 2014–May 2014	78	32.3 (29.4-35.6)	38.6 (35.7-41.8)	0.84 (0.78-0.90)
Summer	Jun. 2014–August. 2014	235	23.4 (21.8-25.1)	16.1 (15.2-17.1)	1.45 (1.38-1.53)
Rush hour					
Rush	(am7-9) and (pm18-20)	584	26.7 (25.4-28.0)	23.7; (22.6-24.8)	1.13 (1.09-1.17)
Non-rush	Excluding rush hours	110	22.4 (20.1, 24.9)	21.4; (19.4-23.6)	1.05 (0.97-1.13)
Diurnal					
Daytime	Hour: 06-17	465	26.8 (25.4-28.3)	23.0 (21.8-24.2)	1.17 (1.12-1.21)
Nighttime	Hour: 18-05	229	24.3 (22.5-26.2)	23.9 (22.3-25.6)	1.01 (0.96-1.08)
Region					
Urban	Dense residential area	330	27.0 (25.3-28.7)	22.3 (20.9-23.7)	1.21 (1.16-1.26)
Industry	Factory area	248	26.7 (24.8-28.7)	24.4 (22.8-26.2)	1.09 (1.03-1.16)
Harbor	Taichung harbor area	116	21.9 (19.5, 24.5)	23.9 (21.4, 26.6)	0.92 (0.84-1.00)
Whole	_	694	25.9 (24.8-27.1)	23.3 (22.3-24.3)	1.11 (1.08-1.15)

TABLE 4 | Comparison of PM<sub>2.5</sub> concentrations of the whole sampling year, seasonal, diurnal, and rush/non-rush hour for data obtained from both the AQMS and MMS.

<sup>a</sup>Ratio of MMS/AQMS = exp (In\_MMS-In\_AQMS).

the AQMS (GM = 23.3  $\mu$ g/m<sup>3</sup>; 95% CI = 22.3–24.3  $\mu$ g/m<sup>3</sup>) (t-test; *p* < 0.01). The former is 1.23 times (95% CI = 1.18-1.27) higher in magnitude than the latter. The aforementioned result is not so surprising since the MMS was much closer to PM<sub>2.5</sub> emission sources (such as traffic) than the AQMS (Wu et al., 2014). In addition, our results are also consistent with previous studies. For example, a study conducted in Boston's Chinatown (near a highway) shows that the increase of building elevation from 0 to 35 m would result in a decrease of the PM2.5 mass concentration by 5.1% (Wu et al., 2014). Another study conducted in Pittsburgh, Pennsylvania, shows that the annual MMS is higher than the annual average of the AQMS (14.6 and 10.2  $\mu$ g/m<sup>3</sup>, respectively) (Li et al., 2016). Here, it should be noted that the latter study was simply conducted in the 2013 winter and 2014 summer (i.e., does not cover four seasons), and hence no statistical significance could be found.

For seasonal PM<sub>2.5</sub> mass concentrations, MMS results are statistically significantly higher than those of the AQMS in both summer (GM = 23.4 and 16.1  $\mu$ g/m<sup>3</sup>, 95% CI = 21.8–25.1 and 15.2–17.1  $\mu$ g/m<sup>3</sup>, respectively; paired t-test; p < 0.001) and winter (GM = 33.1 and 29.3  $\mu$ g/m<sup>3</sup>, 95% CI = 30.3–36.2 and 27.1–31.7  $\mu$ g/m<sup>3</sup>, respectively; paired t-test; p < 0.001). However, an opposite trend is found in both spring (GM = 32.3 and 38.6  $\mu$ g/m<sup>3</sup>, 95% CI = 29.4–35.6 and 35.7–41.8  $\mu$ g/m<sup>3</sup>, respectively; paired t-test; p < 0.001) and fall (GM = 18.5 and 22.3  $\mu$ g/m<sup>3</sup>, 95% CI = 17.3–19.8 and 21.0, 23.6  $\mu$ g/m<sup>3</sup>, respectively; paired t-test; p < 0.001). The opposite trend found in spring and fall might be associated with the lower atmospheric boundary layer, and more significant temperature inversions were found in these two seasons (Ye et al., 2018; Yousefian et al., 2020).

Since the MMS was more proximal to traffic PM<sub>2.5</sub> emission sources than the AQMS, as expected, we found that the MMS rush hour results (GM =  $26.7 \ \mu g/m^3$ , 95% CI:  $25.4-28.0 \ \mu g/m^3$ )

are significantly higher than those of the AQMS (GM =  $23.7 \,\mu g/m^3$ , 95% CI =  $22.6-24.8 \,\mu g/m^3$ ) (paired sample t-test; p < 0.001). Although the same trend can also be found for data collected during non-rush hours, however, no statistical significance is found in the present study (GM = 22.4 and 21.4  $\mu$ g/m<sup>3</sup>, 95% CI = 20.1–24.9 and 19.4–23.6  $\mu$ g/  $m^3$  for the MMS and AQMS, respectively; paired t-test; p =0.234). Obviously, the aforementioned result could be due to density during non-rush hours. less traffic The aforementioned reason can also be used to explain the diurnal sampling results. Here, the daytime sampling results show that those of the MMS are significantly higher than those of the AQMS (GM = 26.8 and 23.0  $\mu$ g/m<sup>3</sup>, 95% CI = 25.4–28.3 and 21.8–24.2  $\mu$ g/m<sup>3</sup>, respectively; paired t-test; p < 0.001)). Although nighttime sampling results for the MMS are higher than those of the AQMS, no statistical significance is found in the present study (GM = 24.3 and 23.9  $\mu$ g/m<sup>3</sup>, 95% CI = 22.5–26.2 and 22.3–25.6  $\mu$ g/m<sup>3</sup>, respectively; paired t-test; p = 0.635).

### Comparisons of PM<sub>2.5</sub> Concentrations of the Urban, Industry, and Harbor Regions Obtained From the MMS and Those Simultaneously Obtained From the AQMS

**Table 4** shows the PM<sub>2.5</sub> concentrations of the urban, industry, and harbor regions, and the whole Shalu area data obtained from the MMS. No significant difference (ANOVA post hoc test, p = 0.978) could be found between PM<sub>2.5</sub> concentrations of the urban region (GM = 27.0 µg/m<sup>3</sup>, 95% CI = 25.3–28.7 µg/m<sup>3</sup>) and industry region (GM = 26.7 µg/m<sup>3</sup>, 95% CI = 24.8–28.7 µg/m<sup>3</sup>). Considering that the dimension of the whole Shalu area is less than 100 km<sup>2</sup>, the geographical adjacency of both regions might lead to a similarity in their

 $PM_{2.5}$  concentrations. On the other hand, both  $PM_{2.5}$  concentrations of the aforementioned two regions were significantly higher than those of the harbor region (GM =  $21.9 \ \mu g/m^3$ , 95% CI =  $19.5-24.5 \ \mu g/m^3$ )  $ug/m^3$  (ANOVA post hoc test; p < 0.01). It could be due to fewer ground-level pollution sources in the harbor region than that in others. In addition, its remote location from the other two regions and the effect of the monsoon wind direction could also be considered contributors (Cheng and Hsu, 2019).

For the comparison purpose, PM<sub>2.5</sub> concentrations of the urban, industry, and harbor regions obtained from the MMS were compared with those simultaneously obtained from the AQMS. For both urban and industrial regions, the MMS concentration (GM = 27.0 and 26.7  $\mu$ g/m<sup>3</sup>, 95% CI = 25.3-28.7 and 24.8–28.7  $\mu$ g/m<sup>3</sup>, respectively) is significantly higher than that of the AQMS (GM = 22.3 and 24.4  $\mu$ g/m<sup>3</sup>, 95% CI = 20.9-23.7 and 22.8–26.2  $\mu$ g/m<sup>3</sup>, respectively) (paired t-test; p <0.001). For the harbor region, however, the MMS concentration (GM =  $21.9 \,\mu g/m^3$ , 95% CI =  $19.5-24.5 \,\mu g/m^3$ )  $m^3$ ) is significantly less than that of the AQMS (GM =  $23.9 \,\mu g/m^3$ , 95% CI = 21.4–26.6  $\mu g/m^3$ ) (paired t-test; p =0.046). The aforementioned results might be because of the existence of fewer ground-level pollution sources in the harbor region. For both urban and industrial regions, the PM<sub>2.5</sub> concentration of the MMS is respectively 1.21 and 1.09 times higher in magnitude than that of the AQMS. The aforementioned results might be because the urban region is proximal to traffic pollution sources, and the industrial region is mainly affected by pollution sources associated with manufacturing and heavy truck emission.

# Establishing the Relationship Between PM<sub>2.5</sub> Concentrations of the AQMS and MMS

Multivariate linear regression (MLR) analyses (hierarchical regression) were conducted to identify variables used for establishing predicting models through examining the variability and collinearity of candidate interference factors. The statistical significance can be found for the four factors, CO, SO<sub>2</sub>, WS, and AQMS PM<sub>2.5</sub> concentrations. The aforementioned four factors were first included for establishing predicting models ( $R^2 = 0.58$ ). Considering that the total contribution of SO<sub>2</sub>, CO, and WS to  $R^2$  is less than 7%, they were removed from the prediction reasons models pragmatically for practical (see Supplementary Table S1). The ignorance of the aforementioned factors is consistent with previous studies conducted in Lubbock, Texas (Kelley et al., 2020), Tianjin, Beijing, and Hebei provinces in China (Zhang et al., 2018). Considering the intrinsic difference in PM<sub>2.5</sub> concentrations of the urban, industrial, and harbor regions, predicting models were established not only based on the data collected from the whole area but also each individual region. The resultant predicting models are shown as follows: Whole area:

$$ln_MMS = 0.839 + 0.768*ln_AQMS, (RMSE = 1.50\mu g/m^3, R^2)$$
$$= 0.54; n = 694).$$
(3)

Urban region:

$$ln_MMS = 0.691 + 0.839*ln_AQMS, (RMSE = 1.42\mu g/m^3, R^2)$$
$$= 0.65; n = 330).$$
(4)

Industrial region:

$$ln_MMS = 1.037 + 0.703*ln_AQMS, (RMSE = 1.55\mu g/m^3, R^2 = 0.45; n = 248).$$
(5)

Harbor region:

$$ln_MMS = 0.714 + 0.747*ln_AQMS, (RMSE = 1.55\mu g/m^3, R^2 = 0.51; n = 116).$$
(6)

Figures 3A-D show the scatter plots of the MMS and AQMS for data collected from the whole study area, urban, industrial, and harbor regions, respectively. The  $R^2$  obtained from the present study is comparable with another study for predicting outdoor personal exposures using nearby AQMS PM2.5 concentrations (Miller et al., 2019). In the present study, the ANOVA (post hoc test) was performed to examine if there is a statistical significance among the four predicting models (See Supplementary Table S2). Results show that no significant difference can be found among the three predicting models for the whole area, urban region, and industrial region; however, the slope of the predicting model for the harbor region was significantly lower than that of others (all p < 0.05). The aforementioned results are consistent with a study conducted in Guangzhou, China, where the relationship between AQMS PM<sub>2.5</sub> concentrations and personal exposure varies greatly from district to district (Jahn et al., 2013). Here, it should be noted that it would be more reasonable to have one predicting model for both urban and industrial regions from a statistical point of view. However, considering the intrinsic difference in the emission sources of the aforementioned two regions, using two separate predicting models could be feasible to meet practical purposes. In the present study, the established four predicting models were adopted for predicting the residents' exposures and further for conducting health impact analyses of each individual region and the whole area.

### Health Impact Assessment for Each Individual Region and the Whole Area

Seven-year (2013–2019) AQMS-measured concentrations (i.e., AQMS<sub>7-yr</sub>) were used to establish the long-term  $PM_{2.5}$  concentrations at the ground level (i.e.,  $MMS_{7-yr}$ ) of the whole area and the three different regions using **Eqs 3–6**, respectively (**Table 5**). Health impact functions and local health data were used to quantify the  $PM_{2.5}$ -attributable health burden for both AQMS<sub>7-yr</sub> and  $MMS_{7-yr}$ , respectively. The above ground-level



 $PM_{2.5}$  concentrations together with the parameter values shown in **Table 3** for lung cancer were applied to the GEMM (**Eq. 1**) for deriving the  $PM_{2.5}$  mortality hazard ratio (HR). Since the mortality probability of lung cancer is small, the HR obtained from the present study was used as a surrogate for the relative risk (RR) (Burnett et al., 2018). **Table 5** also shows the lung cancerattributed fraction (AF) of the population using **Eq. 2**. Results show that both AQMS<sub>7-yr</sub> and MMS<sub>7-yr</sub> decreased from 2013–2019, and the former is lower than the latter. As a result, the same downward trend can also be seen in the estimated RR and AF over the years.

For the whole area, AQMS  $PM_{2.5}$  concentrations from 2013–2019 are, in total, decreased by 56% (i.e., from 35.4 to 15.4  $\mu$ g/m<sup>3</sup>), the resultant decrease for the RR was 20% (i.e., from 1.45 to 1.19) and AF was 48% (i.e., from 0.31 to 0.16). Therefore, reducing the AQMS PM<sub>2.5</sub> concentrations (i.e., AQMS<sub>7-yr</sub>) has a high contribution to the lung cancer AF. Here, it should be noted that even if the yearly average AQMS PM<sub>2.5</sub> concentration meets the government and WHO PM<sub>2.5</sub> interim targets 3 and 4 (i.e., 15 and 10  $\mu$ g/m3, respectively) (W.H.O., 2021), the resultant AFs still cannot be ignored (= 16 and 11%, respectively). Similar results can also be seen in MMS<sub>7-yr</sub> [i.e., PM<sub>2.5</sub> concentrations

decreased by 47% in total (i.e., from 35.8 to  $18.9 \,\mu$ g/m<sup>3</sup>), RR by 15% (i.e., from 1.46 to 1.24), and AF by 39% (i.e., from 0.31 to 0.19)]. Even if the yearly average AQMS PM<sub>2.5</sub> concentration meets the government and WHO PM<sub>2.5</sub> interim targets 3 and 4, the estimated MMS<sub>7-y</sub> (i.e., 18.5 and 13.6, respectively) would lead to even higher AFs (=19 and 14%, respectively) in comparison with the corresponding values (= 16 and 11%, respectively) associated with AQMS PM<sub>2.5</sub> concentrations.

**Table 5** also shows the estimated annual mean of  $PM_{2.5}$  concentrations at the ground level (MMS<sub>7-yr</sub>) of the urban, industrial, and harbor regions, and the corresponding attributed fraction of the population (AF) for lung cancer. The trend in the decrease of  $PM_{2.5}$  concentrations from 2013 to 2019 and the decrease in the resultant RR are similar to those found in the whole area. Moreover, it can also be seen that if AQMS<sub>7-yr</sub> is used for estimating the AF for residents in the Shalu area, an average underestimation of 10.4% was yielded in comparison with those using  $MMS_{7-yr}$ . The decrease of unit  $PM_{2.5}$  concentration would lead to the decrease in the AF of lung cancer by 0.75 and 0.71% in AQMS<sub>7-yr</sub> and  $MMS_{7-yr}$ , respectively. By applying **Eq. 3** to the whole Taiwan area, directly using AQMS<sub>7-yr</sub> would lead to an underestimation of

**TABLE 5** Annual mean of PM<sub>2.5</sub> concentrations of AQMS<sub>7-yr</sub> and the ground level (MMS7-yr) of the whole Shalu area and their derived relative risks (RRs) and the attributed fraction of the population (AF).

	Year						WHO interim target <sup>d</sup>		
	2013	2014	2015	2016	2017	2018	2019	Target 3	Target 4
PM <sub>2.5</sub> (µg/M <sup>3</sup> )	)								
AQMS <sub>7-yr</sub> <sup>a</sup> MMS <sub>7-yr</sub> <sup>b</sup>	35.4	28.7	19.3	21.5	19.3	18.9	15.4	15	10
Whole area	35.8	30.5	22.5	24.4	22.5	22.1	18.9	18.5	13.6
Urban	39.8	33.4	23.9	26.2	23.9	23.5	19.8	19.4	13.8
Industry	34.6	29.9	22.6	24.4	22.6	22.3	19.3	18.9	14.2
Harbor	29.3	25.1	18.6	20.2	18.6	18.3	15.7	15.4	11.4
RR (95%Cl) <sup>c</sup>									
AQMS	1.45	1.37	1.24	1.27	1.24	1.24	1.19	1.19	1.12
	(1.25, 1.69)	(1.2, 1.55)	(1.14, 1.36)	(1.15, 1.4)	(1.14, 1.36)	(1.13, 1.35)	(1.11, 1.28)	(1.11, 1.27)	(1.07, 1.17)
Whole area	1.46	1.39	1.28	1.31	1.28	1.28	1.24	1.23	1.17
	(1.25, 1.7)	(1.21, 1.59)	(1.16, 1.42)	(1.17, 1.46)	(1.16, 1.42)	(1.16, 1.42)	(1.13, 1.35)	(1.13, 1.34)	(1.1, 1.25)
Urban	1.51	1.43	1.3	1.33	1.3	1.3	1.25	1.24	1.17
	(1.28, 1.79)	(1.23, 1.65)	(1.17, 1.45)	(1.18, 1.5)	(1.17, 1.45)	(1.17, 1.44)	(1.14, 1.37)	(1.14, 1.36)	(1.1, 1.25)
Industry	1.44	1.38	1.29	1.31	1.29	1.28	1.24	1.24	1.18
	(1.24, 1.68)	(1.21, 1.58)	(1.16, 1.43)	(1.17, 1.46)	(1.16, 1.43)	(1.16, 1.42)	(1.14, 1.36)	(1.13, 1.35)	(1.1, 1.26)
Harbor	1.37	1.32	1.23	1.26	1.23	1.23	1.2	1.19	1.14
	(1.21, 1.56)	(1.18, 1.48)	(1.13, 1.35)	(1.14, 1.38)	(1.13, 1.35)	(1.13, 1.34)	(1.11, 1.29)	(1.11, 1.28)	(1.08, 1.2)
AF (95% CI)									
AQMS	0.31	0.27	0.2	0.21	0.2	0.19	0.16	0.16	0.11
	(0.2, 0.41)	(0.17, 0.36)	(0.12, 0.26)	(0.13, 0.29)	(0.12, 0.26)	(0.12, 0.26)	(0.1, 0.22)	(0.1, 0.22)	(0.07, 0.15)
Whole area	0.31	0.28	0.22	0.24	0.22	0.22	0.19	0.19	0.14
	(0.2, 0.41)	(0.18, 0.37)	(0.14, 0.3)	(0.15, 0.32)	(0.14, 0.3)	(0.14, 0.29)	(0.12, 0.26)	(0.12, 0.26)	(0.09, 0.2)
Urban	0.34	0.3	0.23	0.25	0.23	0.23	0.2	0.2	0.15
	(0.22, 0.44)	(0.19, 0.39)	(0.14, 0.31)	(0.16, 0.33)	(0.14, 0.31)	(0.14, 0.31)	(0.12, 0.27)	(0.12, 0.27)	(0.09, 0.2)
Industry	0.31	0.28	0.22	0.24	0.22	0.22	0.2	0.19	0.15
-	(0.19, 0.4)	(0.17, 0.37)	(0.14, 0.3)	(0.15, 0.32)	(0.14, 0.3)	(0.14, 0.3)	(0.12, 0.26)	(0.12, 0.26)	(0.09, 0.21)
Harbor	0.27	0.24	0.19	0.2	0.19	0.19	0.16	0.16	0.12
'	(0.17, 0.36)	(0.15, 0.32)	(0.12, 0.26)	(0.13, 0.27)	(0.12, 0.26)	(0.12, 0.25)	(0.1, 0.22)	(0.1, 0.22)	(0.07, 0.17)

<sup>a</sup>The annual average concentration of PM<sub>2.5</sub> (a large amount of data is assumed to be normal distribution) and the data are from the official annual report of TEPA (Environmental Protection Administration, T.R.O.C, 2021).

<sup>b</sup>Regression prediction.

<sup>c</sup>GEMM prediction.

<sup>d</sup>WHO global air quality guidelines (2021) (W.H.O., 2021).

~ 1,000 lung cancer deaths annually in Taiwan during the period from 2013 to 2019 in comparison with those using  $MMS_{7-yr}$  (See **Supplementary Figure S3**). The aforementioned results further strengthen the importance of developing methodologies for predicting  $PM_{2.5}$  concentrations at the ground level.

#### **Limitations and Future Implications**

Considering that there were annual increases in the number of residents, vehicles, and manufacturing industries from 2014 to 2019, it would be more reasonable to establish predicting models based on data collected from the same period of time. In the present study, although before the establishment of the predicting models, possible interference factors were identified (including atmospheric variables, primary pollutants, and secondary pollutants), which would lead to the developed predicting models becoming more suitable for complicated pollution scenarios. Nevertheless, the establishment of the prediction model in the present study is simply based on data collected from 2013 to 2014 for practical reasons. The aforementioned factor should be considered a limitation of the present study. Moreover, possible interference factors in the present study were considered as those affecting PM2.5 concentrations. The selection of possible interference factors was based on the results of literature reviews and available information collected by the AQMS. Indeed, some VOCs are known to be associated with the formation of secondary aerosols, which will also affect the concentrations of PM2.5. However, considering no VOC information could be obtained from the AQMS, only three atmospheric variables, three primary pollutants, and one secondary pollutant were selected. The aforementioned deficiency might explain the  $R^2$  of the predicting models falling to the range 0.45-0.65. We believe that the addition of some VOCs might increase the magnitude of  $R^2$  of the predicting models. The aforementioned factor can also be considered a limitation of the present study.

This study was set out to develop an integrated approach for effectively predicting  $PM_{2.5}$  exposures for residents at the ground level *via* the combination of the concentrations measured across different seasons and regions using the MMS and those obtained

from the nearby fixed-site AQMS. Our results strongly suggest that using the ground-level concentrations would be more accurate in assessing residents' health impacts than those directly obtained from the AQMS. Since the AQMS has been intensively installed in many countries, AQMS data have been widely used for pollution alerts for residents, policy-making for pollution control strategies, and global disease burden assessment. Therefore, the methodology developed from the present study would provide us with a feasible approach for predicting the ground-level concentrations which will effectively enhance the value of the AQMS installed in many countries. Moreover, the approach developed from the present study will be also beneficial to the epidemiological research field for better assessing health outcomes associated with residents'  $PM_{2.5}$  exposures.

## CONCLUSION

In this study, we presented a methodology for assessing the PM<sub>2.5</sub> exposures of residents and their health impacts at a city level. We found that there is a significant linear relationship between MMSmeasured ground-level concentrations and the corresponding concentrations obtained from the AQMS of the area, and the former is higher than that of the latter. A consistent trend in PM2.5 exposures associated with the spatial difference was found as industry region  $\cong$  urban region > harbor region. Our results clearly indicate that residents' ambient PM2.5 exposures do exist with spatial differences in both vertical and horizontal spaces. The health impact assessment results show that the use of AQMS data would lead to an underestimation of ~ 10.4% in magnitude in the estimated lung cancer death attributed fraction (AF) in ground-level comparison with those using exposure concentrations. Similarly, directly using the former would lead

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to an underestimation of ~ 1,000 lung cancer deaths annually in Taiwan during the period from 2013 to 2019, in comparison with those using the latter. The present study clearly indicates the importance of developing methodologies for predicting residents' ground-level  $PM_{2.5}$  exposure concentrations in conducting exposure and health impact assessments.

#### DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

#### **AUTHOR CONTRIBUTIONS**

Study design: P-JT and M-YL. Field data collection: M-SH, M-YL, L-HY, H-TH. Statistical analyses: M-SH, B-FH, and J-DW. Manuscript preparation: M-SH and P-JT.

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#### SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvs.2022.901754/full#supplementary-material

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