Chemical Characteristics, Spatiotemporal Distribution, and Source Apportionment of PM_{2.5} Surrounding Industrial Complexes in Southern Kaohsiung

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ABSTRACT

This study investigated PM_{2.5}, specifically, its chemical characteristics and spatiotemporal variation, and identified its potential sources in the Linhai industrial complex of southern Kaohsiung. Seasonal 12-h PM_{2.5} samples were collected simultaneously at three sampling sites. The results showed that high PM_{2.5} concentrations mainly occurred during winter and spring, with concentrations at the downwind sites always exceeding those at the upwind sites due to the transport of secondary aerosol by the prevailing winds. 31.4–56.8% of the PM_{2.5} consisted of water-soluble ions (WSIs), which were dominated by secondary inorganic aerosol (SIA) ($\text{SO}_4^{2-} + \text{NO}_3^- + \text{NH}_4^+$). High mass ratios between the SIA and the PM_{2.5} (SIA/PM_{2.5}) were also observed during winter and spring, suggesting that SIA was the primary contributor to the high levels of PM_{2.5}. A neutralization ratio (NR) < 1 indicated that the PM_{2.5} was acidic, and a nitrogen oxidation ratio (NOR) > 0.1 and a sulfur oxidation ratio (SOR) > 0.25 showed that SIA frequently formed during winter and spring. The metallic elements, which accounted for 12.0–20.2% of the PM_{2.5}, tended to exhibit higher concentrations during daytime than nighttime, and the enrichment factors (EFs) revealed that the trace metals (Ni, Cr, Cu, and Zn) were mainly anthropogenic in origin. Carbonaceous content formed 9.3–24.3% of the PM_{2.5}, and high mass ratios between the organic and the elemental carbon (OC/EC) were also observed during winter and spring. Moreover, the mass ratios between the malonic and the succinic acid (M/S) were always > 1.0 during winter and spring and < 1.0 during summer and fall, demonstrating that organic acids dominated the SOA during the first two seasons. The major sources of PM_{2.5} in the Linhai industrial complex were steel plants, followed by secondary sulfate and nitrate, vehicular exhaust, petrochemical plants, and incinerators.

Keywords: Industrial complex; Fine particles (PM_{2.5}); Spatiotemporal variation; Chemical characteristics; Source apportionment.

INTRODUCTION

PM_{2.5} pollution has been a hot issue nationwide due to the severe threat to human health. Long-term exposure to PM_{2.5} could cause severe impacts on human health since it is highly suspected as carcinogenic, mutagenic, and teratogenic, which make PM_{2.5} the Type I carcinogenic pollutant issued by World Health Organization (WHO). Additionally, PM_{2.5} could highly possibly penetrate alveoli in the lower respiratory tract of human lungs (Sacks *et al.*, 2010; Christopher *et al.*, 2018) and enter the cardiovascular system (Loop *et al.*, 2018) particularly in metropolitan and industrial areas (Kim *et al.*, 1999; Khan *et al.*, 2010; Lee *et al.*, 2018). Thus,

the improvement of poor particulate air quality has been one of the hottest environmental tasks nationwide and worldwide.

As the biggest industrial city in southern Taiwan, Kaohsiung suffers from the highest concentrations of criteria air pollutants, particularly PM_{2.5}, among the major cities in Taiwan in the past two decades, due to the most densely distributed heavy industries. During 2016, 26.3% of the whole year suffered from severe PM_{2.5} pollution that was much higher than daily ambient air quality standard of 35 $\mu\text{g m}^{-3}$ ruled by the Taiwan Environmental Protection Administration (TWEPA), even higher than 15 $\mu\text{g m}^{-3}$ of yearly standard. Thus, the exploration of the spatiotemporal distribution of PM_{2.5}, as well as its chemical characteristic, has attracted much attention, aiming to depict its chemical fingerprint and track the major source. Yang *et al.* (2017) indicated that the dominant source of PM_{2.5} at the KAQZ was secondary inorganic aerosols, traffic exhausts, and road dust in winter and spring seasons. Shen *et al.* (2019) also investigated the chemical characteristic of PM_{2.5} in the urban, petrochemical

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industry, rural region of Kaohsiung City and Pingtung County, respectively, during highly polluted event days. The identified dominant sources included anthropogenic emissions such as ironworks, coal-fired power plants, petrochemical industry, etc. Since the contribution of industrial emission to primary PM_{2.5} and gaseous precursors (i.e., SO₂, NO_x) of PM_{2.5} plays a key role in the formation of PM_{2.5}, it is crucial to investigate the spatiotemporal distribution and chemical characteristics of PM_{2.5} surrounding the anthropogenic source in Kaohsiung for understanding the fingerprint of PM_{2.5} in highly polluted regions.

The Linhai industrial complex, as the heavy industrial center in southern Taiwan, has approximately 56% of Taiwan's heavy industries with the most prominent infrastructures, including the key industries of ironworks, oil refineries, petrochemical plants, utility power plants, freight transport, and shipbuilding. Additionally, Kaohsiung Linhai Harbor is the busiest seaport with the largest cargo-loading capacity in Taiwan, while the nearby Kaohsiung International Airport is the second busiest airport in Taiwan in terms of passenger numbers. Kaohsiung Linhai Harbor is well connected to other major cities by national highways. However, the systematical investigation on the spatiotemporal distribution and chemical fingerprint of PM_{2.5} surrounding the Linhai industrial complex are lacking in the past years, which is imperative to conduct for forming relevant strategies to eliminate the PM_{2.5} pollution.

The influence of prevailing winds on the spatiotemporal distribution of PM_{2.5} in southern Kaohsiung is significant. Previous studies reported that higher PM_{2.5} concentrations are mostly observed in winter (northeasterly winds) because higher concentration of PM_{2.5} could be blown from Northeast Asia, North Asia, and even Central Taiwan in winter and spring, promoting and aggregating the local ambient PM_{2.5} concentration (Sacks *et al.*, 2010; Loop *et al.*, 2018). By contrast, clean air carried by southwesterly winds in summer influence relatively less on the formation of PM_{2.5} in Taiwan (Li *et al.*, 2017). Additionally, the formation of secondary PM_{2.5} may exhibit significant diurnal variation due to the sunlight radiation; anthropogenic emission of SO₂, NO_x, volatile organic compounds (VOCs), etc. diurnally varied as well, thereby leading to the diurnal pattern of concentration and chemical characteristic of PM_{2.5} (Chen *et al.*, 2019). Therefore, it is critical to investigate the seasonal and diurnal variation of PM_{2.5} concentration and their chemical fingerprint.

Accordingly, this study aims to investigate the chemical characteristics, the spatiotemporal variation of PM_{2.5} surrounding the largest industrial complexes of southern Kaohsiung. The seasonal and diurnal variations of PM_{2.5}'s chemical composition, including water-soluble ions (WSIs), metallic elements, anhydrosugars, and organic acids, were thoroughly explored. Finally, the seasonal and diurnal sources of PM_{2.5} and their contributions were identified using backward trajectories, principle component analysis (PCA), and the receptor model of chemical mass balance (CMB).

METHODOLOGIES

Sampling Protocol

Seasonal 24-h PM_{2.5} were collected simultaneously at

three sampling sites (i.e., National Open University, Fongyang Elementary School, and Fonglin Elementary School) surrounding the Linhai industrial complex in southern Kaohsiung (Fig. 1). The Linhai industrial complex is the largest industrial complex in Taiwan, including ironworks, coal-fired power plants, oil refinery, and petrochemical plants. Among these three sampling sites, National Open University site (NOU) is located at the southern boundary of Kaohsiung International Airport (KIA), and is approximately 20 m above the ground. Fongyang Elementary School site (FY) is close to Kaohsiung City Library and National Kaohsiung University of Hospitality and Tourism (NKUHT), and is approximately 15 m above the ground. Fonglin Elementary School site (FL) is located nearby Dalin refining plant of China Petroleum Company (CPC), Dalin coal-fired power plant of Taiwan Power Company (TPC), China Steel Company (CSC), and Kaohsiung Harbor, and is approximately 15 m above the ground.

This study applied PM_{2.5} samplers (PQ-200, BGI) to collect 12-h PM_{2.5} samples at a flow rate of 16.67 L min⁻¹ from December 2017 to September 2018 in different seasons. Four PM_{2.5} sampling protocols were conducted on December 4–8 in 2017 (winter samples), March 12–16 (spring samples), July 10–13 (summer samples), and November 17–21 in 2018 (fall samples), respectively.

Quartz fibrous filters of 47 mm were used in the PM_{2.5} samplers for collecting 12-h PM_{2.5} for consecutive five days in each season. Daytime PM_{2.5} sampling started from 07:00 to 19:00, while nighttime PM_{2.5} sampling started from 07:00 to 19:00 (next day). After sampling, PM_{2.5} filters were then transported back to Air Pollution Laboratory in the Institute of Environmental Engineering at National Sun Yat-sen University as soon as possible and temporarily conditioned in a thermostatic chamber with a constant temperature (T) of 20–25°C and a relative humidity (RH) of 40 ± 5% for at least 48 h for weighing by an analytical microbalance with the precision of 1 µg (MSA6.6S, Sartorius) to determine the mass concentration of PM_{2.5}.

Chemical Analytical Methods

After weighing, PM_{2.5} filters were divided into four identical portions for further chemical analysis. One quarter of the quartz fibrous filter was used for analyzing the water-soluble ions, which was put into a 50 mL PE bottle poured with 30 mL distilled deionized water (D.I. H₂O) with conductivity > 18 MΩ to dissolve ionic species into water from PM_{2.5} by an ultrasonic vibrator for approximately 120 min. An ion chromatography (IC) (DX-120, Dionex) was then used to analyze the concentrations of major anions (F⁻, Br⁻, Cl⁻, SO₄²⁻, and NO₃⁻) with 1.8 mM Na₂CO₃/1.7 mM NaHCO₃ solution as an eluent. Another ion chromatography (ICS-1100, Dionex) was further used to analyze the concentrations of major cations (NH₄⁺, K⁺, Na⁺, Ca²⁺, and Mg²⁺) with 20 mM methane sulfonic acid as an eluent.

Another quarter of the quartz fibrous filter was used for analyzing metallic content, which was initially digested in a 20 mL mixed acid solution (HNO₃:HClO₃ = 3:7) at a temperature of 150–200°C for 120 min, and then diluted to 25 mL with D.I. H₂O. During the digestion process, D.I.

Fig. 1. Location of three PM_{2.5} sampling sites surrounding the Linhai industrial complex in southern Kaohsiung.

H₂O was added to the residual solution at least two times in order to eliminate the acidic content of the digestion solution. The metallic elements in PM_{2.5}, including Cr, Mn, Fe, Ni, Zn, Cd, Pb, Mg, K, Ca, Ti, Al, As, V, Cu, Ba, and Sb, were then analyzed with an inductively coupled plasma/atomic emission spectrometry (ICP/AES) (Optima 2000 DV, PerkinElmer).

The carbonaceous contents of PM_{2.5} were analyzed with an elemental analyzer (EA) (Vario EL III, Elementar). One quarter of the quartz fibrous filter was cut into two identical parts. One eighth of the quartz fibrous filter was preheated in advance by hot nitrogen gas (340–345°C) for 30 min to expel the organic carbon (OC) fraction, after which the amount of elemental carbon (EC) was then determined. Another eighth of the quartz fibrous filter was analyzed without preheating, and the carbon content was thus characterized as total carbon (TC). OC was further determined by extracting EC from TC.

Another quarter of the quartz fibrous filter was analyzed for anhydrosugars (i.e., levoglucosan, mannosan, and galactosan) and organic acids (i.e., oxalic acid, malonic acid, and succinic acid) with a high-performance ionic chromatography (HPIC) (ICS-5000+, Dionex). The quartz fibrous filters were initially extracted with the distilled deionized water with conductivity > 18.2 MΩ in a PE bottle under ultrasonic vibration for 120 min with a prebaked quartz fibrous filter (0.3 μm pore size). Anhydrosugars were analyzed by using 200, 100, 18, and 5 mM NaOH as the eluents at a flow rate of 1 mL min⁻¹.

Organic acids were analyzed by using 25 mM H₂SO₄ and 5 mM NaOH as the eluents at a flow rate of 1 mL min⁻¹.

Backward Trajectory Simulation

In order to trace air masses transported toward the sampling sites, backward trajectories were plotted by using the wind field including wind direction and wind speed obtained from local air quality monitoring stations and weather stations set up by Taiwan Environmental Protection Administration (TEPA) and Taiwan Central Weather Bureau (TCWB), respectively. In this mode, the starting time of backward trajectory simulation was selected as the occurrence time of the highest PM_{2.5} concentration during the sampling period, and traced back by the airflow trajectory till 24:00 in the current day. As shown in the output maps, 1 h was used as the time interval of the track data.

Principal Component Analysis (PCA)

Principal component analysis is one of the methods of multivariate statistical analysis, which has been widely applied to resolve the main loading factors in sequence for characterizing the principal components with larger variances. The principle of PCA uses the obtained eigenvalues and eigenvectors to screen for a larger variance of each component (Turias *et al.*, 2006; Han *et al.*, 2015). The level-of-correlation loading factor is shown below:

- (1) High correlation—loading factor higher than 0.7,
- (2) Moderate correlation—loading factor between 0.5–0.7, and
- (3) Low correlation—loading factor less than 0.5.

The principal component analysis applied chemical analytical data to determine the main loading factors. The largest loading factor is the first principal component, and followed by the sequence of the second, the third, and the fourth principal components. However, the main purpose of the principal component analysis was to apply less variance to represent big data, commonly simplify to 3–4 main loading factors.

Chemical Mass Balance (CMB) Receptor Model

The source apportionment of $PM_{2.5}$ was resolved by using a chemical mass balance receptor model based on the principle of mass conservation. A CMB Version 8.0 model was applied by verifying the following fit indices, such as χ^2 (≤ 4.0), R^2 (≥ 0.8), the percentage of mass (80–120%), and T statistics (≥ 2.0). The residual and uncertainty ratios were required between -2.0 and 2.0 , and their values should be as close as zero, in order to obtain better analytical results. The higher degrees of freedom, the better the resolution of CMB analysis. In this study, the source apportionment of $PM_{2.5}$ was carried out by using CMB Version 8.0 model. Comprehensive knowledge of the physical and chemical characteristics of fine particles ($PM_{2.5}$) measured at both the source and the receptor sites are required to identify the types of $PM_{2.5}$ sources and quantify their contributions at the receptor site. The relationship between the concentrations of chemical species measured at the receptor site and those emitted from the sources can be presented by Eq. (1):

$$C_i = \sum_{j=1}^p \alpha_{ij} F_{ij} S_j \quad (1)$$

where C_i is the concentration of species i measured at the receptor site ($\mu\text{g m}^{-3}$). α_{ij} is the coefficient of species i generated or sunk during the transportation process. F_{ij} is the mass fraction of species i in the profile of the source j (%). S_j is the mass concentration of all species at the receptor site assigned to the source j ($\mu\text{g m}^{-3}$).

RESULTS AND DISCUSSION

Spatiotemporal Variation of $PM_{2.5}$ Concentration

Fig. 2 illustrates the seasonal and diurnal variation of $PM_{2.5}$ concentration measured at three sampling sites in the Linhai industrial complex. The average $PM_{2.5}$ concentration at National Open University sampled in winter, spring, summer, and fall were 41.4 , 38.5 , 12.0 , and $16.8 \mu\text{g m}^{-3}$, respectively. While, those at Fongyang Elementary School were 30.1 , 33.3 , 19.1 , and $29.0 \mu\text{g m}^{-3}$, respectively, and those at Fonglin Elementary School were 38.6 , 27.8 , 13.2 , and $25.4 \mu\text{g m}^{-3}$, respectively. In addition, although the concentrations in summer and fall nearly approached the yearly standard of $15 \mu\text{g m}^{-3}$ by Taiwan Environmental Protection Agency, those were still far higher than the yearly average $PM_{2.5}$ concentration standard of $10 \mu\text{g m}^{-3}$ ruled by WHO, as illustrated in Fig. 2.

The correlation coefficient of manual sampling (three $PM_{2.5}$ sampling sites of this study) and automatic sampling (three adjacent TEPA air quality stations) results was as high as 0.98.

In terms of spatial distribution, NOU had the highest average $PM_{2.5}$ concentration in winter and spring with the prevailing northeastern winds, and FY has the highest average $PM_{2.5}$ concentration in summer and fall with the prevailing southern winds, implying that NOU and FY located at the downwind sites of $PM_{2.5}$ sources were highly affected by local sources in the Linhai industrial complex.

The diurnal variation of $PM_{2.5}$ concentrations was highly influenced by the synoptic-scale meteorology and anthropogenic activities. This study found that nighttime

Fig. 2. Seasonal and diurnal variation of $PM_{2.5}$ concentration at three sampling sites in southern Kaohsiung.

PM_{2.5} concentrations were mostly higher than those in the daytime at NOU and FY, but an opposite trend was observed at FL, in spring and summer, implying that sampling sites located at the downwind of the Linhai industrial complex had higher PM_{2.5} concentrations at nighttime. On the contrary, daytime PM_{2.5} concentrations were always higher than those at nighttime in fall and winter. It is speculated that the three sampling sites were greatly influenced by human activities in the daytime in fall and winter.

In order to further understand the spatiotemporal variation of PM_{2.5} in the Linhai industrial complex, this study also conducted 24-h sampling of PM_{2.5} in four seasons simultaneously at ten sampling sites, including National Open University (NOU), Fongyang Elementary School (FY), Fonglin Elementary School (FL), China Steel Aluminum Corporation (CSAC), Hongmaogang Cultural Park (HMG), Linyuan Elementary School (LY), Sanwei Elementary School (SW), Kangshi Elementary School (KS), Xixi Village Activity Center (XX), and Chinese Petroleum Corporation (CPC). The measured PM_{2.5} concentrations were then used to plot the seasonal contour of PM_{2.5} concentration in southern Kaohsiung. The contour maps of PM_{2.5} covering two neighboring industrial complexes in four seasons are illustrated in Fig. 3.

Two local maximum of PM_{2.5} concentrations were observed at HMG (98.0 µg m⁻³) and CPC (51.2 µg m⁻³) in winter while blowing prevailing northeastern winds. High PM_{2.5} concentration at HMG was mainly attributed to ship exhausts at the main entrance of Kaohsiung Harbor, while CPC was located at the downwind site of two industrial complexes. Overall, the spatiotemporal distribution of PM_{2.5} concentrations clearly showed a trend of PM_{2.5} concentration at the downwind sites were always higher than those at the upwind sites, indicating that prevailing winds could blow both primary and secondary aerosols from the upwind region to the downwind sites.

Chemical Characteristics of PM_{2.5}

Water-soluble Ion Analysis

In this study, PM_{2.5} samples collected at National Open University, Fongyang Elementary School, and Fonglin Elementary School in four seasons were further analyzed for five chemical contents, including water-soluble ions, metallic elements, carbonaceous contents, anhydrosugars, and organic acids.

As illustrated in Fig. 4, WSIs accounted for 31.8–58.4%, 31.1–56.1%, 32.1–55.6% of PM_{2.5} sampled at NOU, FY, and FL, respectively. For all PM_{2.5} samples, the most abundant ionic species in PM_{2.5} were SO₄²⁻, NO₃⁻, and NH₄⁺, namely secondary inorganic aerosol (SIA). The seasonal variations of WSIs and PM_{2.5} mass concentration were very much similar during the sampling period. Moreover, the concentrations of SO₄²⁻, NO₃⁻, and NH₄⁺ had a tendency to rise significantly since fall and winter. Overall speaking, SIA accounted for 66.0–82.9% of WSIs in PM_{2.5} at all sampling sites (Table 1). As far as the seasonal variation of SIA, high concentrations of SIA were observed in winter at three sampling sites, while low concentrations of SIA were found in summer. The highest SIA concentration of 15.1 µg m⁻³

was observed at NOU at nighttime, while the lowest SIA concentration of 3.19 µg m⁻³ was observed at FL in the daytime. It implied that the particulate air quality of southern Kaohsiung was deteriorated mainly by ammonium sulfate ((NH₄)₂SO₄) and ammonium nitrate (NH₄NO₃), which were formed in the atmosphere by SO₂, NO_x, and NH₃ emitted from local sources and/or long-range transport. Similar to PM_{2.5} concentration, SIA in PM_{2.5} sampled at the downwind sites were commonly higher than those at the upwind sites, implying that air contaminants tended to accumulate in the downwind region and thus increase the concentrations of PM_{2.5}.

In order to further ascertain the chemical characteristics of PM_{2.5} in the Linhai industrial complex, the equivalent concentrations of SO₄²⁻, NO₃⁻, and NH₄⁺ in PM_{2.5} were used to determine the neutralization rate (NR), sulfur oxidation rate (SOR), and nitrogen oxidation rate (NOR). The neutralization ratios (NRs) of PM_{2.5} which represents the aerosol acidity originating from particulate NO₃⁻ and SO₄²⁻, were firstly evaluated to analyze the acidity of PM_{2.5} using the following equations:

$$NR = \frac{[NH_4^+]}{S_{nss-SO_4^{2-}} + [NO_3^-]} \quad (2)$$

The SOR and NOR as the sulfur and nitrogen oxidation ratios are described by the following equations:

$$SOR = \frac{S_{nss-SO_4^{2-}}}{S_{nss-SO_4^{2-}} + S_{SO_2}} \quad (3)$$

$$NOR = \frac{S_{nss-NO_3^-}}{S_{nss-NO_3^-} + S_{NO_2}} \quad (4)$$

where nss-SO₄²⁻ is the excess non-sea-salt sulfate, calculated by subtracting the amount of SO₄²⁻ in seawater from that of SO₄²⁻ in fine atmospheric particles; nss-NO₃⁻ is the excess non-sea-salt nitrogen, calculated by subtracting the amount of NO₃⁻ in seawater from that of NO₃⁻ in PM_{2.5}.

The diurnal variation of NR, SOR, and NOR of PM_{2.5} at three sampling sites in four seasons are summarized in Table 2. It clearly showed that the average NRs were always < 1.0 in all seasons, indicating that NH₄⁺ cannot solely neutralize nss-SO₄²⁻ and NO₃⁻ in PM_{2.5}. In these cases, PM_{2.5} in southern Kaohsiung was thought of as acid particles since water-soluble ions were dominated by acidic species (e.g., SO₄²⁻ and NO₃⁻) during the sampling periods in all seasons.

Moreover, the average SORs were mostly > 0.25 in winter and spring, while those in summer and fall were always < 0.25. However, different results were found for NORs, showing that the average NORs were always > 0.1 in all seasons. Therefore, PM_{2.5} was dominated by both secondary sulfate and nitrate in winter and spring, resulting in the deterioration of particulate air quality. It suggested that secondary sulfate and nitrate could be converted from SO₂ and NO₂ via

Fig. 3. Seasonal variation of $PM_{2.5}$ concentration contour in southern Kaohsiung.

Fig. 4. Seasonal and diurnal variation of water-soluble ions in $PM_{2.5}$ at three sampling sites in southern Kaohsiung.

Table 1. Seasonal average of SIA, SIA/WSIs, chloride deficits, and $[Cl^-]/[Na^+]$ of $PM_{2.5}$ at three sampling sites in southern Kaohsiung.

	Sampling Sites	Winter	Spring	Summer	Fall
SIA ($\mu g m^{-3}$)	NOU-D	15.1	10.2	3.7	6.3
	NOU-N	14.5	17.7	4.5	5.7
	FY-D	11.3	10.2	4.3	11.7
	FY-N	10.5	13.7	5.7	10.6
	FL-D	11.7	10.3	3.2	10.1
	FL-N	14.3	11.5	3.4	9.0
SIA/WSIs (%)	NOU-D	78.1	71.8	68.7	81.9
	NOU-N	79.9	77.5	75.4	79.1
	FY-D	82.8	74.3	67.4	75.9
	FY-N	80.9	78.9	71.4	78.5
	FL-D	75.9	76.2	67.8	79.4
	FL-N	78.4	78.1	65.9	75.3
Chloride Deficit (%)	NOU-D	48.8	50.9	44.9	42.6
	NOU-N	53.0	50.7	44.5	45.8
	FY-D	44.6	53.4	47.2	45.5
	FY-N	49.5	52.9	47.0	47.0
	FL-D	47.8	49.4	47.8	46.8
	FL-N	47.0	51.5	47.1	43.9
$[Cl^-]/[Na^+]$ Molar Ratio	NOU-D	0.92	0.89	1.00	1.03
	NOU-N	0.85	0.89	1.00	0.97
	FY-D	1.00	0.89	1.02	0.98
	FY-N	0.91	0.85	0.95	0.95
	FL-D	0.94	0.91	0.94	0.96
	FL-N	0.95	0.87	0.95	1.01

NOU: National Open University; FY: Fongyang Elementary School; FL: Fonglin Elementary School; D: Daytime; N: Nighttime; SIA: Secondary inorganic aerosol ($= [NH_4^+] + [SO_4^{2-}] + [NO_3^-]$); WSIs: Water-soluble ions.

atmospheric chemical reactions and transported to southern Kaohsiung during the northeastern monsoon periods. During the winter and spring days, sulfate and nitrate existing in the atmosphere had pretty much high potential to react with ammonium and form secondary ammonium sulfate and ammonium nitrate, also known as secondary inorganic aerosols which could be formed chemically by homogeneous reaction (mainly gas-gas), heterogeneous reaction (including

gas-solid and gas-liquid), and photochemical reaction in the atmosphere and thus increased the amount of SIA in $PM_{2.5}$ during the winter and spring days.

As shown in Table 1, the $[Cl^-]/[Na^+]$ molar ratios ranged from 0.85 to 1.03, while the chloride deficit ranged from 43.9 to 53.4%, during the sampling periods, suggesting that $PM_{2.5}$ was less affected by the oceanic spray. Such a phenomenon was mainly attributed to the fact that the $PM_{2.5}$

Table 2. Seasonal variation of NR, SOR, and NOR of PM_{2.5} at three sampling sites in southern Kaohsiung.

Seasons	Sampling Sites		NR	SOR	NOR
Winter	NOU	D	0.648	0.403	0.124
		N	0.840	0.384	0.135
	FY	D	0.985	0.300	0.094
		N	0.825	0.329	0.099
	FL	D	0.903	0.217	0.089
		N	1.050	0.222	0.115
Spring	NOU	D	0.835	0.299	0.094
		N	0.848	0.548	0.180
	FY	D	0.866	0.312	0.110
		N	0.816	0.475	0.250
	FL	D	1.032	0.172	0.107
		N	1.053	0.148	0.089
Summer	NOU	D	0.595	0.089	0.090
		N	0.715	0.129	0.105
	FY	D	0.784	0.131	0.134
		N	0.879	0.211	0.209
	FL	D	0.547	0.067	0.207
		N	0.667	0.038	0.177
Fall	NOU	D	0.779	0.186	0.096
		N	0.673	0.218	0.117
	FY	D	0.631	0.229	0.244
		N	0.864	0.281	0.250
	FL	D	0.826	0.271	0.222
		N	0.869	0.140	0.171

NOU: National Open University; FY: Fongyang Elementary School; FL: Fonglin Elementary School; D: Daytime; N: Nighttime.

sampling sites were located at the inland region of southern Kaohsiung. Moreover, the chloride deficit had a negative correlation with $[\text{Cl}^-]/[\text{Na}^+]$ molar ratio, indicating that the $[\text{Cl}^-]/[\text{Na}^+]$ molar ratio tended to decrease as the chloride deficit increased. Previous researches reported that the mass ratio of $[\text{Cl}^-]/[\text{Na}^+]$ is about 1.8 (or the molar ratio of 1.17) of sea salts in the oceanic spray mainly due to low chloride deficit (Cheng *et al.*, 2000; Seinfeld and Pandis, 2006). Chloride could react with acidic substances (e.g., HNO_3 , H_2SO_4 , and other acids) and form HCl which is then evaporated to the atmosphere. The mass ratio of $[\text{Cl}^-]/[\text{Na}^+]$ is thus reduced gradually while sea salts migrate toward inland in the atmosphere due to chloride deficit. The chloride deficit can be determined by the following Eqs. (5) and (6) (Kerminen *et al.*, 1998):

$$\text{Chloride Deficit (\%)} = ([\text{Cl}^-]_{\text{original}} - [\text{Cl}^-]) / [\text{Cl}^-]_{\text{original}} \times 100\% \quad (5)$$

$$[\text{Cl}^-]_{\text{original}} = [\text{Na}^+] \times ([\text{Cl}^-]/[\text{Na}^+])_{\text{original}} \quad (6)$$

In terms of seasonal variation, the chloride deficits of PM_{2.5} at National Open University, Fongyang Elementary School, and Fonglin Elementary School in winter and spring were consistently higher than those in summer and fall (Table 1). The results indicated that chloride deficit was correlated to East Asian monsoons, mainly due to high atmospheric concentrations of acidic gases in southern Kaohsiung during the northeastern monsoon seasons (Yuan

et al., 2017, 2018). On the contrary, in summer and fall, the southwestern monsoons blow relatively clean air masses from the seas to the sampling sites, resulting in lower atmospheric concentrations of acidic gases and thus lower chloride deficit.

Metallic Element Analysis

Fig. 5 illustrates the seasonal and diurnal variation of metallic element concentrations in PM_{2.5} sampled at three sampling sites in southern Kaohsiung. It showed that crustal elements (Ca, K, Mg, Fe, and Al) were the dominant metallic elements in PM_{2.5}. The seasonal variation of metallic elements had the same trend as PM_{2.5} concentration. In terms of spatial distribution, the concentrations of metallic elements at National Open University were generally higher than those at Fongyang Elementary School and Fonglin Elementary School. Additionally, trace elements (Ti, V, Cr, Mn, Ni, Cu, As, Pb, and Cd) emitted mainly from anthropogenic sources were lower than crustal elements in all seasons.

Diurnal variation results showed that daytime metallic elements in PM_{2.5} were mostly higher than those at nighttime particularly in winter and spring, while an opposite trend was observed in summer and fall. The metallic contents of PM_{2.5} were dominated by crustal materials (Ca, Fe, Al, and Ti), oceanic sprays (Mg and K), anthropogenic sources (Cd, V, Cu, Cr, Pb, and As), and mobile sources (Zn), as shown in Fig. 5. The diurnal variation of Mg, K, Ca, V, Fe, Cu, Zn, Al, As, Cd, and Pb mainly emitted from crustal materials, oceanic sprays, and vehicular exhausts were insignificant.

Fig. 5. Seasonal and diurnal variation of metallic elements in $PM_{2.5}$ at three sampling sites in southern Kaohsiung.

However, a significant diurnal variation of trace elements (Ti, Cr, Mn, and Ni) was observed, suggesting that the anthropogenic emission from industrial process influenced the metallic component of $PM_{2.5}$ (Shen *et al.*, 2019).

This study further ascertains the characteristics of metallic elements in $PM_{2.5}$ by using enrichment factor (EF), which is defined as the relative abundance of metallic elements to reference element to explore the correlation between $PM_{2.5}$ and crustal materials at the sampling sites. In the present study, calcium (Ca) as the most abundant crustal metal in $PM_{2.5}$ was used as the reference element to determine EF (Yuan *et al.*, 2006) and identify whether $PM_{2.5}$ was dominated by crustal materials or anthropogenic sources (Chester *et al.*, 2009). If $EF > 10$, metallic elements in $PM_{2.5}$ were contributed mainly from anthropogenic sources. If $EF < 10$, metallic elements in $PM_{2.5}$ were derived mainly from crustal materials.

The enrichment factors of metallic elements in $PM_{2.5}$ sampled at the sampling sites are depicted in Fig. 6. It showed that the EF_s of Ni and Cr elements were mostly > 10 in all seasons except for fall, while the EF_s of Cu and Zn were mostly < 10 in all seasons except for spring. The results indicated that trace metals, including Ni, Cr, Cu, and Zn, came mainly from anthropogenic sources. Previous literature reported that Ni and Cr came mainly from ironworks, metal processing, and coal burning, while Cu and Zn came mainly from smelters and metal processing. Unlike Ni, Cr, Cu, and Zn, the EF_s of Al, Ti, Mg, Fe, Pb, K, and Ba were always < 10 , indicating that these metallic elements were derived mainly from crustal sources. Overall speaking, the most severe metallic pollution contributed to $PM_{2.5}$ was observed in spring, while the lowest one occurred in fall.

Carbonaceous Content Analysis

The seasonal and diurnal variation of carbonaceous content

of $PM_{2.5}$ sampled at three sampling sites is illustrated in Fig. 7. Similarly, the diurnal variation of carbonaceous contents and mass concentrations of $PM_{2.5}$ were quite similar. It showed that the concentrations of OC in $PM_{2.5}$ were always higher than those of EC at three sampling sites in all seasons. Among them, EC mainly came from inert carbonaceous materials, diesel engines, fuel combustion, industrial combustion, and exhaust gases (Kim *et al.*, 1999). Elemental carbon has a chemical structure similar to impure graphite, originating primarily from direct emissions of combustion (Chen *et al.*, 2016). Furthermore, the mass ratios of organic carbon and elemental carbon (OC/EC) in winter and spring were always higher than those in summer and fall (Fig. 7). Since fall, the OC/EC mass ratios increased gradually at all sampling sites. This study further used the minimum of OC/EC mass ratios ($(OC/EC)_{min}$) (see Fig. 8) to estimate the secondary organic carbon (SOC) in $PM_{2.5}$ (Cao *et al.*, 2004). The concentration of SOC can be determined by either Eq. (7) or (8) (Kerminen *et al.*, 1998):

$$SOC = OC - EC \times (OC/EC)_{pri} \quad (7)$$

$$SOC = OC - EC \times (OC/EC)_{min} \quad (8)$$

Herein, the SOC of $PM_{2.5}$ can be obtained by subtracting primary organic carbon, $(EC) \times (OC/EC)_{pri}$, from OC. However, in reality, $(OC/EC)_{pri}$ in Eq. (4) was not available, thus $(EC) \times (OC/EC)_{min}$ was deemed as primary organic carbon (POC) in this study (Wu *et al.*, 2009). In practice, the $(OC/EC)_{min}$ is readily obtained from the minimum slope of the OC versus EC, as illustrated in Fig. 8.

The seasonal and diurnal variation of estimated SOC concentrations in $PM_{2.5}$ at three sampling sites in southern Kaohsiung is illustrated in Fig. 9. It showed that the concentrations of SOC in winter and spring were commonly

Fig. 6. Seasonal and diurnal variation of enrichment factor (EF) of $PM_{2.5}$ at three sampling sites in southern Kaohsiung.

higher than those in summer and fall, which caused a severe impact on ambient carbonaceous aerosols along the coastal regions during the northeastern monsoon periods. Elemental carbon could also be generated from atmospheric photochemical reactions during long-range transport, which formed SOC in the atmosphere (Li *et al.*, 2017). In addition to long-range transport, volatile organic compounds emitted from industrial complexes were responsible for the

formation of SOC during the winter days, since atmospheric photochemical reactions could convert VOCs to SOC.

Anhydrosugar Analysis

Anhydrosugars, including levoglucosan, mannosan, and galactose, are the most efficient organic tracers for organic aerosols emitted from biomass burning in the atmosphere. As illustrated in Fig. 10, levoglucosan was the only anhydrosugar

Fig. 7. Seasonal and diurnal variation of carbon concentration and OC/EC ratio in $PM_{2.5}$ at three sampling sites in southern Kaohsiung.

Fig. 8. $(OC/EC)_{min}$ of $PM_{2.5}$ during the sampling periods in southern Kaohsiung.

Fig. 9. Seasonal and diurnal variation of SOC concentration in $PM_{2.5}$ at three sampling sites in southern Kaohsiung.

being detected, while mannosan and galactose were not detectable (ND) in PM_{2.5} in the present study. The average concentrations of levoglucosan in 12-h PM_{2.5} samples at all sampling sites ranged from 30.3 to 55.3 ng m⁻³. High concentrations of levoglucosan in the range of 47.2–54.9 ng m⁻³ were observed mainly in spring, showing that biomass burning was one of the dominant contributors to atmospheric PM_{2.5}, which concurred with the more frequent occurrence of biomass burning in spring than other seasons in the Indochinese Peninsula, as shown in Fig. S1 (Liu *et al.*, 2013; Bhattarai *et al.*, 2019). Moreover, no apparent diurnal variation trend of levoglucosan concentration was observed at all sampling sites in four seasons. In terms of spatial distribution, the concentrations of levoglucosan were ordered as NOU > FL > FY in spring since the polluted air mass was blown northerly from northern Taiwan and/or long-range transported from southeastern and northeastern Asia. However, an opposite trend was observed in the other three seasons (Fig. 10).

Organic Acid Analysis

The seasonal and diurnal variation of organic acids in

PM_{2.5} at three sampling sites in southern Kaohsiung is depicted in Fig. 11. It showed that the seasonal variation of organic acid concentrations was similar to PM_{2.5} mass concentrations. Overall, the concentrations of organic acids in winter and spring were commonly higher than those in summer and fall. Among the three measured organic acids, oxalic acid was the most abundant organic acid in PM_{2.5}. The concentrations of oxalic acid at three sampling sites ranged from 176.2 to 647.7 ng m⁻³ in winter and spring. Previous literature reported that the mass ratios of malonic and succinic acids (M/S) have been applied to determine whether atmospheric PM_{2.5} was mainly attributed to secondary organic aerosol (Kawamura *et al.*, 1993). This study revealed a significant seasonal variation of M/S mass ratios. During the sampling periods of winter and spring, the M/S mass ratios of PM_{2.5} ranged from 0.98 to 1.21, which were much higher than those ranging from 0.49 to 0.81 in summer and fall. High M/S mass ratios indicated that PM_{2.5} in winter and spring was highly correlated with secondary organic aerosols that were long-range transported from the far north toward southern Kaohsiung (Tsai *et al.*, 2013; Dhananjay *et al.*, 2019).

Fig. 10. Seasonal and diurnal variation of levoglucosan concentration in PM_{2.5} at three sampling sites in southern Kaohsiung.

Fig. 11. Seasonal and diurnal variation of organic acid concentration in PM_{2.5} at three sampling sites in southern Kaohsiung.

Simulated Backward Trajectories

In the present study, backward trajectories were applied to trace air masses transported toward southern Kaohsiung. Air trajectories were plotted by using Xiaogang air quality monitoring station (located at Xiaogang Junior High School) as the ending point for tracking the backward trajectories (Figs. 12–13). In winter (on December 4–8), air trajectories came mainly from western Taiwan (i.e., Taichung, Yunlin, and Chiayi), which were transported from the north toward the Xiaogang site on December 4–5. However, air trajectories on December 6 and 8 were blown from the Taiwan Strait, turned from the north to the south, and then moved toward the Xiaogang site.

In spring (on March 12–16), air trajectories on March 12–14 came from the Taiwan Strait, passed through southern Tainan, and arrived at southern Kaohsiung, which transported mainly from the north to the south. Air trajectories on March 15 transported from Pingtung County from the western Taiwan Strait, which mainly transported from the east to the west. While air trajectories on March 16 were transported from the northwestern to the southeastern Taiwan Strait.

In summer (on July 10–13), air trajectories were relatively disordered compared to the other three seasons. Air trajectory on July 10 was mainly transported from the northwestern to the southeastern Tainan and then moved toward the Xiaogang site. Air trajectory on July 11 came from the Taiwan Strait, which was blown to the eastern and western Pingtung and then transported toward the Xiaogang site. Air trajectory on July 13 was blown from the northwestern to the southeastern Taiwan Strait.

In fall (on November 17–21), air trajectory on November 17 came from the northeastern to the southwestern Taitung, crossing the Central Range, and then blown downward the Xiaogang site. Air trajectory on November 18 was transported from the north to the south to Pingtung County, and finally blown from the east to the west and moved toward the Xiaogang site. Air trajectory on November 19 was blown mainly from the western coastline of Taiwan and moved from the north to the south. Air trajectories on November 20–21 were mainly transported from the Kaohsiung offshore toward the Xiaogang site, which moved mainly from the west to the east.

Source Identification and Apportionment of PM_{2.5}

The identification and apportionment of PM_{2.5} sources based on PCA at three sampling sites are shown in Table 3. The PCA result showed that the chemical species statistically clustered in each major factor were distinguished in different seasons. In winter, the highest principal component (PC1) contained Mg²⁺, Al, Ti, Pb, and Ba, explaining 37.1% of the total variance, which were closely associated with steel plants or metal processing (Amodio *et al.*, 2013; Zhou *et al.*, 2019). The second principal component (PC2) included NO₃⁻, SO₄²⁻, NH₄⁺, and OC, accounting for 14.1% of the variance, were identified as secondary aerosols. The third and fourth principal components (PC3 and PC4) consisted of Cl⁻, Na⁺, and Mg, Ca were linked to oceanic spray and fugitive dust, respectively. In spring, PC1 with the total variance of 33.4% consisted of NH₄⁺, Ni, Cu, and EC, which

were probably related to coal-burning boilers. PC2 that had V and Pb, explaining 10.9% of the variance, represented metal processing. In addition, V was also probably emitted from coal burning (Pei *et al.*, 2016). PC3 with K and Ba, accounting for 10.2% of the total variance, was probably emitted from steel plants. PC4 with Na⁺ and Cl⁻ stood for ocean sprays. In summer, the PC1 made up of Al, Ti, V, Mn, Pb, Cu, and Ba, accounting for 45.7% of the total variance, was attributed to steel plants or metal processing. PC2 (12.6%) and PC3 (8.3%) containing NH₄⁺, and K, SO₄²⁻, Ca²⁺, and Ca, respectively, both of which were closely related to road dust (Zhang *et al.*, 2018). Na⁺ and Cl⁻ in PC4 (7.1%) mainly originated from the oceanic spray. In fall, PC1 (32.4%) involving OC and EC was directly related to vehicular exhausts (Watson *et al.*, 2001; Corbin *et al.*, 2018; Li *et al.*, 2018). Metallic elements of Pb, Ba, Cr in PC2 (14.8%) were also considered to be emitted from steel plants or metal processes. V, Fe, and Ni in PC3 (13.0%) were considered to be bound in the primary fine particles emitted from fuel oil combustion or vehicular exhausts (Khan *et al.*, 2016; Elhadi *et al.*, 2018). The metallic elements of Ti and Al in PC4 (8.5%) mainly come from road dust. To sum up, the chemical components of PM_{2.5} statistically resolved majorly originated from anthropogenic activities in each season, while that derived from secondary aerosols was only found in the winter season, suggesting that photochemical conversion of the precursors were probably more intense in winter (Shen *et al.*, 2019).

The chemical species screened by PCA were selected for further analysis using the CMB receptor model. The results of resolved prominent sources of PM_{2.5} at three sampling sites showed that the largest source of PM_{2.5} in the Linhai industrial complex was steel plants, accounting for 19.8–26.5% of total PM_{2.5}, suggesting that metal processing, blast furnace, fuel combustion, etc. play a vital role in the emission or formation of PM_{2.5}. Secondary aerosols, as the second largest source, contributed 13.3–17.8% of total PM_{2.5}. It is worth noting that the proportion of secondary sulfates exceeded that of secondary nitrates, revealing that the formation of PM_{2.5} was more influenced by stationary sources than vehicular exhausts in the Linhai industrial complex. Additionally, in summer and fall, the proportion originating from secondary aerosol contribution most, about 13.3–15.4% of PM_{2.5}, while in winter and fall that contributed rose to 16.8–17.8%, suggesting that the northeastern monsoon periods (in winter and spring), long-range transport or poor dispersion condition resulted in the formation of secondary aerosols and thus rose their contribution to PM_{2.5}.

Other anthropogenic sources of petrochemical industries (8.9–13.4%), industrial boilers (9.0–10.7%), vehicular exhausts (7.4–11.8%), and incinerators (6.1–8.3%) also dominated the PM_{2.5} source profile (Fig. 14). Besides the industrial activities, the contribution of biomass burning in winter and spring, contributing 4.4–4.5% of PM_{2.5} was commonly higher than that of 3.2–3.4% in summer and fall, suggesting more agricultural activities in winter and spring than those in summer and fall. To sum up, the PM_{2.5} around the Linhai industrial complex was mainly influenced by primary aerosol emitted from anthropogenic activities, and subsequently by secondary aerosols.

Fig. 12. Backward trajectories of air masses in winter and spring.

Fig. 13. Backward trajectories of air masses in summer and fall.

CONCLUSIONS

This study investigated the seasonal and diurnal variation in the PM_{2.5} concentration and chemical composition in the Linhai industrial complex in southern Kaohsiung. The average concentrations ranged from 27.8 to 31.4 $\mu\text{g m}^{-3}$ during the northeastern monsoon periods of winter and spring but were much lower, between 12.0 and 29.0 $\mu\text{g m}^{-3}$, during summer and fall. The concentrations at the downwind sites always exceeded those at the upwind sites. The SORs were frequently > 0.25 during winter and spring, whereas the NORs were

always > 0.1 regardless of the season. Al, Mg, Fe, Ti, Pb, K, and Ba were the dominant metallic elements in the PM_{2.5}, and the trace elements, Ni, Cr, Cu, and Zn, were mainly emitted from anthropogenic sources. The OC/EC ratios were higher during winter and spring than summer and fall, and the SOC concentration displayed a similar trend. High concentrations of levoglucosan, ranging from 47.2 to 54.9 ng m^{-3} , were observed in winter and spring due to biomass burning during the northeastern monsoon periods, and the organic acids in the PM_{2.5} also significantly increased during these two seasons. The high M/S ratios for the PM_{2.5}, which ranged

Table 3. PCA loading factors of PM_{2.5} at three sampling sites in four seasons in southern Kaohsiung.

Principal Components	Winter		
	Chemical Components	Variance (%)	Accumulation (%)
PC1	Mg ²⁺ , Al, Ti, Pb, Ba	37.1	37.1
PC2	NO ₃ ⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , OC	14.1	51.2
PC3	Cl ⁻ , Na ⁺	9.5	60.7
PC4	Mg, Ca	7.9	68.6
Principal Components	Spring		
	Chemical Components	Variance (%)	Accumulation (%)
PC1	NH ₄ ⁺ , Ni, Cu, EC	33.4	33.4
PC2	V, Pb	10.9	44.3
PC3	K ⁺ , Ba	10.2	54.5
PC4	Na ⁺ , Cl ⁻	7.8	62.3
Principal Components	Summer		
	Chemical Components	Variance (%)	Accumulation (%)
PC1	Al, Ti, V, Mn, Pb, Cu, Ba	45.7	45.7
PC2	NH ₄ ⁺ , K	12.6	58.3
PC3	SO ₄ ²⁻ , Ca ²⁺ , Ca	8.3	66.6
PC4	Cl ⁻ , Na ⁺	7.1	73.7
Principal Components	Fall		
	Chemical Components	Variance (%)	Accumulation (%)
PC1	EC, OC	32.4	32.4
PC2	Pb, Cr, Ba	14.8	47.2
PC3	V, Fe, Ni	13.0	60.2
PC4	Al, Ti	8.5	68.7

PCA: Principal component analysis.

(a) Winter

Fig. 14. Seasonal and diurnal variation of source apportionment of PM_{2.5} at three sampling sites in southern Kaohsiung: (a) winter, (b) spring, (c) summer, and (d) fall. Red characters stand for the secondary sulfate and secondary nitrate.

Fig. 14. Seasonal and diurnal variation of source apportionment of $PM_{2.5}$ at three sampling sites in southern Kaohsiung: (a) winter, (b) spring, (c) summer, and (d) fall. Red characters stand for the secondary sulfate and secondary nitrate.

from 0.98 to 1.21, were attributable to long-range-transported primary and secondary organic compounds. The prominent resolved $PM_{2.5}$ sources in the Linhai industrial complex were steel plants (19.8–26.5%), followed by secondary aerosols (13.3–17.8%), petrochemical industries (8.9–13.4%), industrial boilers (9.0–10.7%), vehicular exhaust (7.4–11.8%),

and incinerators (6.1–8.3%). During the northeastern monsoon periods (in winter and spring), primary pollutants emitted locally and/or long-range transported toward southern Kaohsiung resulted in the formation of secondary inorganic and organic aerosol, thereby increasing their contributions to the $PM_{2.5}$.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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