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Seasonal variation and source apportionment of PM$_{2.5}$-bound trace elements at a coastal area in southwestern Taiwan

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Abstract

The aim of this study is to investigate the seasonal variations and source apportionment on atmospheric fine particulate matter (PM$_{2.5}$) mass and associated trace element concentrations at a coastal area, in Chiayi County of southwestern Taiwan. Particle measurements were conducted in 2015. Twenty-three trace elements in PM$_{2.5}$ were analyzed using inductively coupled plasma mass spectrometry (ICP-MS). Multiple approaches of the enrichment factor (EF) analysis and positive matrix fraction (PMF) model were used to identify potential sources of PM$_{2.5}$-bound trace elements. Daily mean concentration of PM$_{2.5}$ in cold season (25.41 μg m^{-3}) was higher than that in hot season (13.10 μg m^{-3}). The trace elements contributed 11.02 and 10.74% in total PM$_{2.5}$ mass concentrations in cold season and hot season, respectively. The results of EF analysis confirmed that Sb, Mo, and Cd were the top three anthropogenic trace elements in the PM$_{2.5}$; furthermore, carcinogenic elements (Cr, Ni, and As) and other trace elements (Na, K, V, Cu, Zn, Sr, Sn, Ba, and Pb) were attributable to anthropogenic emissions in both cold and hot seasons; however, highly enriched Li and Mn were observed only in cold season. The PMF model identified four main sources: iron and steel industry, soil and road dust, coal combustion, and traffic-related emission. Each of these sources has an annual mean contribution of 8.2, 27.5, 11.2, and 53.1%, respectively, to PM$_{2.5}$. The relative dominance of each identified source varies with changing seasons. The highest contributions occurred in cold season for iron and steel industry (66.2%), in hot season for traffic-related emission (58.4%), soil and road dust (22.0%), and coal combustion (2.8%). These findings revealed that the PM$_{2.5}$ mass concentration, PM$_{2.5}$-bound trace element concentrations, and their contributions were various by seasons.

Keywords Fine particulate matter · Trace elements · Enrichment factor · Source apportionment · Seasonal variation

Introduction

In October 2013, the International Agency for Research on Cancer (IARC) has classified particulate matter (PM), a major component of air pollution, as carcinogenic to humans (group 1), based on sufficient evidence that exposure is associated with an increased risk of lung cancer (IARC 2016). Besides,

the toxicity of PM is related to the compositions which are attached to it, and several of which have been classified by the IARC in the group 1 carcinogens, including polycylic aromatic hydrocarbons (PAHs) and some heavy metals (Straif et al. 2013). Numerous epidemiologic studies have reported that

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both short- and long-term exposure to fine PM (aerodynamic diameter ≤ 2.5 μm, $PM_{2.5}$) have been associated with an increase in morbidity and mortality related to cardio-pulmonary diseases (Dominici et al. 2006; Kloog et al. 2012; Li et al. 2016b; Lu et al. 2015; Hwang et al. 2016; Hwang et al. 2017a; Hwang et al. 2017b; Hwang et al. 2017c). $PM_{2.5}$ is a heterogeneous mixture with various chemical components such as water-soluble ions [sulfate (SO_4^{2-}) and nitrate (NO_3^-)], trace elements, and carbonaceous species (Spurny 1998; Xu et al. 2012). The toxicity and carcinogenicity of $PM_{2.5}$ is based on its oxidative potential, which is relative to $PM_{2.5}$ compositions (Crobeddu et al. 2017). One meta-analysis study reported that both elemental carbon (EC) and secondary inorganic aerosols (SO_4^{2-} and NO_3^-) within the $PM_{2.5}$ were positively associated with increased all-cause, cardiovascular, and respiratory mortality, with the strongest associations observed for EC: 1.30% increase in all-cause mortality per 1 μg m^{-3} (Atkinson et al. 2015). Additionally, animal studies and human historical data analyses have suggested that $PM_{2.5}$-bound trace elements [such as iron (Fe), vanadium (V), nickel (Ni), chromium (Cr), copper (Cu), and zinc (Zn)] with the ability to generate reactive oxygen species (ROS), which represents an important mechanism underlying the $PM_{2.5}$-induced detrimental health effects (Chen and Lippmann 2009; Lippmann and Chen 2009).

The elevated $PM_{2.5}$ concentration has been recognized as an important environmental issue in Taiwan because its concentration consistently exceeds the World Health Organization (WHO) Air Quality Guideline (10 μg m^{-3} for annual mean and 25 μg m^{-3} for 24-h mean) (Environmental Protection Administration Executive Yuan 2016; World Health Organization 2016). The Taiwan Environmental Protection Administration (TWEPA) reported that the highest $PM_{2.5}$ concentration typically occurs in the southwestern Taiwan; moreover, the annual average level of $PM_{2.5}$ in southwestern Taiwan is consistently at least four times higher than the WHO guidelines (Environmental Protection Administration Executive Yuan 2016). Taiwan is located on the southeast fringe of East Asia, and the regional climate is affected by the sea-land breezes (SLBs) and the northeastern monsoon (NEW). SLBs and the NEW could carry particulate pollutants originating from South East Asian continent and China to Taiwan (Tsai et al. 2011; Hwang et al. 2016) and play important roles in the distribution and transport of PM in the atmosphere over the southeastern coastal region of Taiwan Strait. Furthermore, most heavy-polluting industrial plants and industrial parks have been established in western coastal Taiwan, which has been recognized in relation to regional poor air quality (Hsu et al. 2016). Recent epidemiological studies have demonstrated that atmospheric $PM_{2.5}$ is associated with respiratory diseases such as chronic obstructive pulmonary disease and asthma in southwestern Taiwan

(Hwang et al. 2016; Hwang et al. 2017b; Hwang et al. 2017c). Lo et al. (2017) found that in 2014, $PM_{2.5}$ was accounted for 6282 deaths in Taiwan, and substantial geographic variation in population attributable fraction (PAF) was observed, with the southwest Taiwan having the highest PAF. Additionally, Hsu et al. (2016) revealed that the concentrations of carcinogenic elements [nickel (Ni), arsenic (As), and chromium (Cr)] in $PM_{2.5}$ in a rural residence of central Taiwan exceeded the guideline limits published by WHO; moreover, the excess cancer risks (ECRs) via the inhalation for carcinogenic elements in $PM_{2.5}$ was 25.9×10^{-6}. Only limited investigations have been performed to evaluate the characteristics of seasonal variations for atmospheric $PM_{2.5}$-bound trace elements in Taiwan (Tseng et al. 2016; Hsu et al. 2016).

The present study aims to analyze seasonal variations in $PM_{2.5}$ mass concentrations and $PM_{2.5}$-bound trace elements at the coastal area of Chiayi County, in southwestern Taiwan. In addition, the possible sources of $PM_{2.5}$ were identified using multiple approached of the enrichment factor (EF) analysis and positive matrix fraction (PMF) model.

Methods

Site description and sample collection

In this study, the sampling site was located at the coastal area of Chiayi County, in southwestern Taiwan. Sampling site is illustrated in Fig. 1. The sampling station was installed on the roof (9 m above ground) of a building of the Chang Gung University of Science and Technology, Chiayi Campus, codenamed "Puzi station" (23.46 N, 120.27 E). A high-volume sampler (Model 6070, Tisch Environmental, Inc., USA) was used to collect $PM_{2.5}$ samples along with the use of quartz microfiber filter paper (20.3 cm × 25.4 cm, Whatman™, UK). The sampler operated at a flow rate of 1.13 m^3 min^{-1} for 24 h. The samples were collected in cold season (January to April) and hot season (May to September) in 2015. The sampling procedure used followed the standard procedure (NIEA A205.11C) announced by TWEPA. Concentrations of carbon dioxide (CO_2), ozone (O_3), nitrogen dioxide (NO_2), and meteorological parameters including temperature and relative humidity (RH) were detected by the KD Engineering AIRBOXX monitor; moreover, wind speed (WS) was detected by the VelociCheck Air Velocity Meter (Model TSI 9515). A total of 90 samples were collected, covering the time from January 2015 to September 2015.
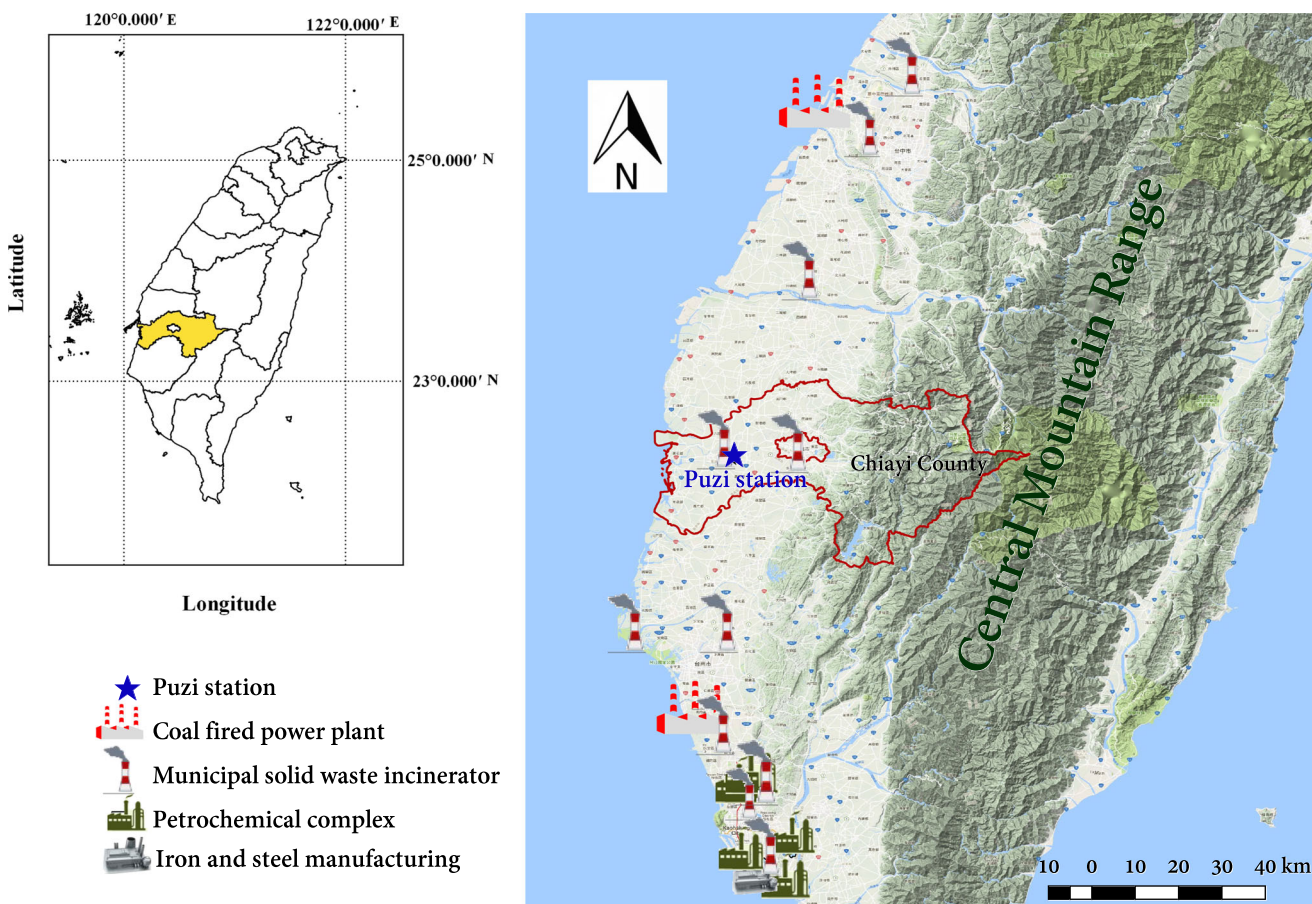
Fig. 1 Location of sampling station: Puzi station (23.46 N, 120.29 E) and selected industrial relevant to the source apportionment presented in this study

Gravimetric and trace elements analysis for PM$_{2.5}$ compositions

Before and after the sampling procedure, the filters were kept for 48 h in desiccators with relative humidity at $40 \pm 5\%$ and temperature at 20 ± 3 °C before being weighted by a microbalance. Each filter sample was weighted at least three times in a row before and after sampling to ensure the variance among measurements for each sample was less than 5 mg. The net mass concentration of PM$_{2.5}$ was obtained by subtracting pre-weight from post-weight.

For analysis of trace elements, samples were acid digested using a mixture solution of 5 mL nitric acid (HNO$_3$) and 0.3 mL hydrofluoric acid (HF) for the first 90 min and 2.8 mL 5% boric acid (H$_3$BO$_3$) for another 60 min by using an ultrahigh-throughput microwave digestion system (Anton-Paar Multiwave 3000). Then, the extracts were cooled and diluted to 15 mL with 5% H$_3$BO$_3$. Extracted solutions were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) with an Agilent-7700× (Agilent Technologies, Santa Clara, CA, USA) instrument. The National Environmental Analysis (NIEA) method M105.01B was used for analysis of 23 trace elements, including lithium (Li), sodium (Na), magnesium

(Mg), aluminum (Al), (K), calcium (Ca), titanium (Ti), V, Cr, manganese (Mn), Fe, cobalt (Co), Ni, Cu, Zn, As, strontium (Sr), molybdenum (Mo), cadmium (Cd), stannum (Sn), antimony (Sb), barium (Ba), and lead (Pb). The method detection limit (MDL) was defined as three times the standard deviation of seven successive blank samples. The MDLs were in the range 1.83×10^{-2}–24.3 μg L^{-1} for all selected trace elements. Accuracy and precision of the instrument were measured using Standard Urban Dust Material (NIST, SRM-2786). One field blank with six to seven field samples was collected for background contamination. The field and filter blanks were analyzed using the same procedure as the samples. There was no significant difference between the filter and field blanks. The blank value of each selected trace elements was subtracted from the analyzed values from the real samples.

Source identification methods

The EF analysis was adopted to indicate the source origin (anthropogenic or crustal) of trace element abundances for atmospheric PM$_{2.5}$. The EF was estimated as the ration of each element's abundance in PM$_{2.5}$ samples to its average

Table 1 Descriptive statistics of meteorological conditions and air pollutant concentrations in cold and hot seasons over the study period in the Puzi station, Taiwan, 2015

Variable	Cold season ($n = 40$)			Hot season ($n = 50$)		
	Mean	SD	Range	Mean	SD	Range
Meteorological conditions						
Temperature (°C)	23.42	3.65	15.85−30.35	29.14	1.35	26.55−32.25
Relative humidity (%)	59.73	8.23	30.60−75.95	74.86	4.83	62.95−83.75
Wind speed (m s^{-1})	1.54	0.80	0.38−3.41	1.51	0.93	0.36−3.54
Pollutant concentrations						
PM$_{2.5}$ (µg m^{-3})	25.41	10.37	6.11−50.87	13.10	7.57	2.45−35.62
CO$_2$ (ppm)	450.83	28.84	375.00−517.00	438.94	15.86	407.00−476.00
O$_3$ (ppb)	98.23	85.91	0.00−362.50	275.39	103.97	30.50−485.00
NO$_2$ (ppm)	0.09	0.04	0.00−0.15	0.12	0.04	0.00−0.20

Cold season: January to April; Hot season: May to September

SD standard deviation

abundance in the upper continental crust (Taylor and McLennan 1995), by selecting Al as the reference element. The EF of trace element in PM$_{2.5}$ sample can be calculated by using the following equation:

$$EF = \frac{\left(\frac{E}{Al}\right)_{PM}}{\left(\frac{E}{Al}\right)_{Crust}}$$

where $(E/Al)_{PM}$ is the concentration ratio of a given trace element E to Al in collected ambient PM$_{2.5}$, and $(E/Al)_{crust}$ is the concentration ratio of the trace element E to Al in the average crust abundance.

Positive Matrix Factorization (PMF v5.0) model (United States Environmental Protection Agency 2014) was employed to identify the contributions of various emission sources. The PMF model, a receptor-based source apportionment model, is a multivariate factor analysis tool that decomposes the matrix of a speciated sample into two matrices: factor contributions and factor profiles (Paatero and Tapper 1994; Paatero 1997). The PMF model calculation is presented by the following equation:

$$X_{ij} = \sum_{k=1}^{P} g_{ik} \cdot f_{kj} + e_{ij}$$

where X_{ij} is the concentration of chemical species j measured on sample i, P is the number of factors contributing to the samples, f_{kj} is the concentration of species j in factor profile k, g_{ik} is the relative contribution of factor k to sample i, and e_{ij} is error of the PMF model for the species j measured on sample i.

The aim of PMF is to identity the values of g_{ik} and that f_{kj} best reproduce x_{ij}. The PMF solution minimizes the objective function Q based on the uncertainties (u) as follows (United States Environmental Protection Agency 2014).

$$Q(E) = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{X_{ij} - \sum_{k=1}^{p} g_{ij} f_{kj}}{u_{ij}} \right]^2$$

where n is the number of samples, m is the number of species, p is the number of sources included in the analysis, and u_{ij} is the uncertainty of ambient concentration of species j in i.

The signal-to-noise ratio (S/N ratio) for each species was calculated, and species were divided into three different groups relating to their S/N ratio. The species were categorized as "strong" if S/N ≥ 2, "weak" if $0.2 \leq$ S/N < 2, and "bad" if S/N < 0.2. In addition, PM$_{2.5}$ mass concentration was included as a "total variable" and conducted PM$_{2.5}$ mass as "weak" in the PMF model to ensure it did not affect the PMF performance.

Statistical analysis

Descriptive statistics (mean, standard deviation, range, and percentage) were used to describe the air pollutant concentrations, meteorological conditions, and PM$_{2.5}$-bound trace elements. Pearson correlation (r) was used to determine correlation between air pollutants and meteorological conditions. The criterion for statistical significance was $p < 0.05$. Statistical analysis was performed using SPSS 20 (IBM Corp., Armonk, NY, USA).

Results

Seasonal variations for ambient pollutant concentration (PM$_{2.5}$, CO$_2$, O$_3$, and NO$_2$) and meteorological variables

Table 1 presents the meteorological conditions (temperature, RH, and WS) and air pollutant concentrations (PM$_{2.5}$, CO$_2$,

Table 2 Pearson coefficients of correlation between air pollutants and meteorological conditions in cold and hot seasons

Cold season

	$PM_{2.5}$	CO_2	O_3	NO_2	Temperature	Relative humidity	Wind speed
$PM_{2.5}$	1.000	0.082	−0.209	0.208	−0.254	−0.214	−0.222
CO_2		1.000	0.118	−0.345*	0.619**	0.126	−0.480**
O_3			1.000	0.362*	0.388*	0.744**	−0.002
NO_2				1.000	−0.520**	0.320*	−0.021
Temperature					1.000	0.280	−0.039
Relative humidity						1.000	0.095
Wind speed							1.000

Hot season

	$PM_{2.5}$	CO_2	O_3	NO_2	Temperature	Relative humidity	Wind speed
$PM_{2.5}$	1.000	0.385**	−0.028	0.135	−0.180	−0.004	−0.424**
CO_2		1.000	−0.474**	−0.337*	0.379**	−0.518**	−0.040
O_3			1.000	0.820**	−0.415**	0.697**	0.092
NO_2				1.000	−0.438**	0.730**	−0.159
Temperature					1.000	−0.721**	0.190
Relative humidity						1.000	−0.227
Wind speed							1.000

*$p < 0.05$; **$p < 0.01$

O_3, and NO_2), on average, obtained from the study area for cold and hot seasons. Generally, $PM_{2.5}$ concentrations show a remarkable seasonal variability with higher level in cold season (25.41 µg m^{-3}) than in hot season (13.10 µg m^{-3}); besides, there was higher level of CO_2 in cold season (CO_2, 450.83 ppm) than in hot season (CO_2, 438.94 ppm). However, there were higher levels of O_3 and NO_2 in hot season than in cold season, with 275.39 µg m^{-3} (O_3) and 0.12 ppm (NO_2) in hot season, and 98.23 µg m^{-3} (O_3) and 0.09 ppm (NO_2) in cold season, respectively. On the other hand, the mean value of temperature, RH, and WS in cold was 23.42 °C, 59.73%, and 1.54 m s^{-1} in cold season and 29.14 °C, 78.86%, and 1.51 m s^{-1} in hot season. Correlations between the air pollutants and meteorological conditions are listed in Table 2. Significant positive correlation between $PM_{2.5}$ and CO_2 ($r = 0.385$, $p < 0.01$) was observed only in hot season. On the other hand, significant negative correlation between $PM_{2.5}$ and WS ($r = -0.424$, $p < 0.01$) was observed only in hot season.

Seasonal variations for trace elemental compositions

Table 3 indicates the mean concentrations of trace elements in $PM_{2.5}$ in cold and hot seasons. In cold season, in addition to Na and K (more than 100 ng m^{-3}), Fe, Mg, Ca, Zn, Al, and Ba (10–100 ng m^{-3}) were also major trace elements in the $PM_{2.5}$, followed by Mn, Cu, Pb, Sr, and V (1–10 ng m^{-3}). The rest of elements with concentrations less than 1 ng m^{-3} were Ti, Ni, As, Mo, Cr, Sn, Sb, Cd, Li, and Co. In hot season, Na and K with a high concentration (more than 100 ng m^{-3}) were observed, while Fe, Ca, Mg, Al, and Ba had moderate abundance in concentrations (10–100 ng m^{-3}). Zn, Cu, Mn, Pb, Sr, and V were at even lower concentrations (1–10 ng m^{-3}), while Ti, Ni, Cr, Mo, As, Sn, Sb, Cd, Li, and Co with the lowest concentrations (less than 1 ng m^{-3}) were observed. To sum up, the trace elements contributed 11.02% (in cold season) and 10.74% (in hot season) of the total $PM_{2.5}$ mass concentrations. Seasonally, the percentage of $PM_{2.5}$-bound trace elements for some elements (Na, Mg, Al, K, Mn, Zn, As, and Ba) were higher in cold season than those in hot season.

Enrichment factor of trace elements

Figure 2 illustrates the results of EF analysis for $PM_{2.5}$-bound trace elements in cold and hot seasons. An EF higher than 10 is considered to indicate that elements are significantly predominantly originate from human activities (Chen et al. 2015; Birmili et al. 2006). The top three anthropogenic elements (Sb, Mo, and Cd) with high EFs in the $PM_{2.5}$ were observed. Other trace elements (Na, K, V, Cr, Ni, Cu, Zn, As, Sr, Sn, Ba, and Pb) had EFs higher than 10 in both cold and hot seasons, indicating that these elements may originate from anthropogenic origins. However, trace elements (Li and Mn) had EFs

Table 3 Summary statistics of PM$_{2.5}$-bound trace elements in cold and hot seasons

Species	Cold season ($n = 40$)			Hot season ($n = 50$)		
	Mean (ng m^{-3})	SD	Trace elements compositions in PM$_{2.5}$ (%)	Mean (ng m^{-3})	SD	Trace elements compositions in PM$_{2.5}$ (%)
Li	0.10	0.06	0.0004	0.10	0.03	0.0004
Na	1708.06	642.16	7.9439	1772.90	423.78	7.8208
Mg	46.32	28.60	0.1877	42.08	10.98	0.1834
Al	23.22	15.08	0.1825	39.19	24.49	0.1800
k	484.21	276.12	1.9270	419.00	136.57	1.7849
Ca	31.52	9.72	0.1650	47.74	12.56	0.2122
Ti	0.80	0.51	0.0032	0.98	0.32	0.0043
V	1.35	0.57	0.0062	1.73	0.30	0.0080
Cr	0.55	0.15	0.0027	0.87	0.62	0.0036
Mn	4.96	3.24	0.0189	3.90	1.30	0.0165
Fe	66.08	50.69	0.2377	69.92	23.89	0.2954
Co	0.06	0.04	0.0002	0.05	0.02	0.0002
Ni	0.78	0.32	0.0035	0.90	0.26	0.0040
Cu	3.92	1.52	0.0176	5.17	2.77	0.0222
Zn	24.66	33.92	0.1562	6.00	12.11	0.0435
As	0.74	0.45	0.0029	0.61	0.24	0.0026
Sr	1.78	0.55	0.0085	2.13	0.70	0.0093
Mo	0.58	0.11	0.0034	0.69	0.31	0.0035
Cd	0.11	0.07	0.0004	0.11	0.06	0.0004
Sn	0.45	0.33	0.0016	0.60	0.36	0.0024
Sb	0.32	0.19	0.0012	0.31	0.13	0.0013
Ba	16.29	17.04	0.1380	30.56	25.15	0.1261
Pb	2.36	1.77	0.0085	2.70	1.61	0.0108
Total			11.02			10.74

higher than 10 only in cold season, suggesting that they may originate from anthropogenic origins in cold season.

Source apportionment

The annual and seasonal source contributions to PM$_{2.5}$-bound trace elements are illustrated in Figs. 3, 4, and 5. The PMF results showed that traffic-related emission, soil and road dust, coal combustion, and iron and steel industry were the main contributors at the coastal area in Chiayi County of southwestern Taiwan. Annually, the first source of trace elements of PM$_{2.5}$ is considered as traffic-related emission (53.1%) because of the high content of V (40.9%), Mn (38.1%), and Ni (38.0%). The second source of trace elements is related to soil and road dust (27.5%) due to the high content of Ca (50.9%), Cu (42.2%), V (36.5%), Ni (30.8%), Pb (30.4%), and Ti (28.5%). The third source of trace elements is considered as coal combustion (11.2%) because of the high content of Pb (48.5%), Mn (37.6%), Cu (36.6%), and Fe (32.9%). The fourth source of trace elements is associated with iron and

steel industry (8.2%) due to the high content of V (40.9%), Mn (38.1%), and Ni (38.0%).

In cold season, iron and steel industry (66.2%) was the predominant contribution to PM$_{2.5}$-bound trace elements because of the high content of Ca (26.2%), Ti (20.5%), Pb (15.9%), and Fe (15.8%). The second source of trace elements is related to traffic-related emission (33.8%) due to the high content of V (55.9%), Ni (47.9%), Mn (39.5%), and Cu (38.8%). As for hot season, the first source of trace elements in PM$_{2.5}$ is considered as traffic-related emission (58.4%) because of the high content of V (30.4%), Cu (23.4%), and Ni (21.0%). The second source of trace elements is related to soil and road dust (22.0%) due to the high content of Ca (44.9%), Ti (42.6%), Ni (41.1%), Mn (38.9%), V (38.5%), and Fe (37.3%). The third source of trace elements is considered as iron and steel industry (16.8%) because of the high content of Ti (40.6%), Cu (31.9%), Ca (25.1%), Pb (23.3%), and Fe (21.9%). The fourth source of trace elements is associated with to coal combustion (2.8%) due to the high content of Pb (63.1%), Mn (50.2%), Cu (44.7%), and Fe (40.7%).
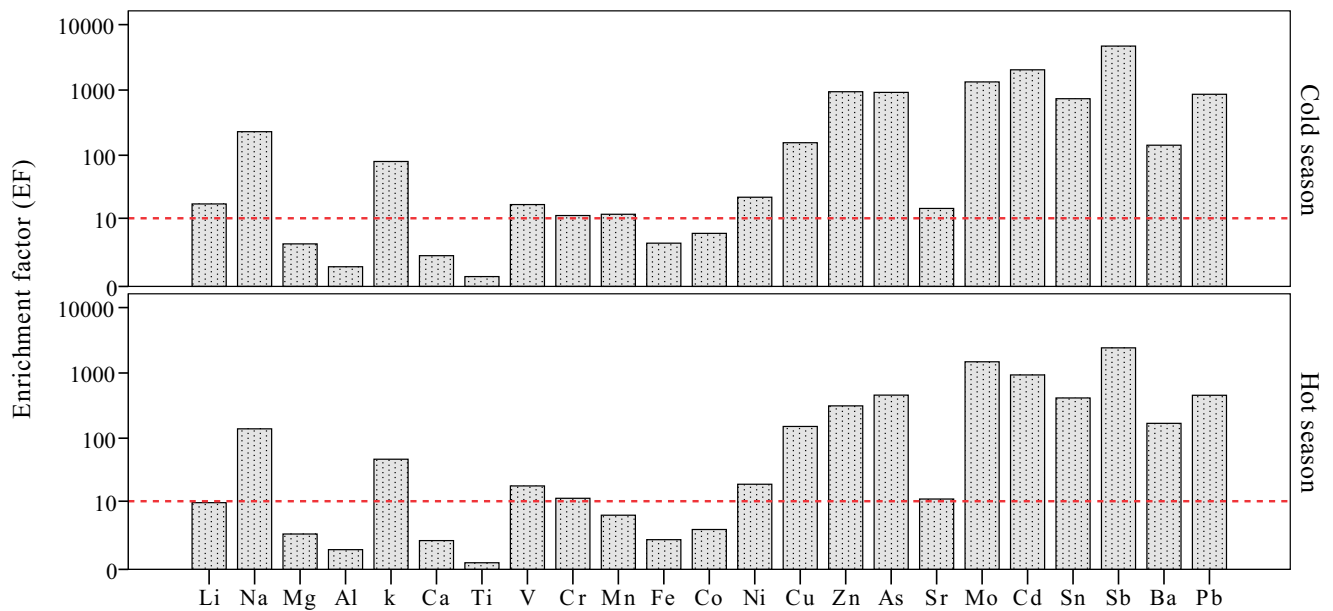
Fig. 2 The EF values for trace elements in PM$_{2.5}$ obtained from cold and hot seasons

Discussion

This study investigated the seasonal variation and sources apportionment of PM$_{2.5}$ and PM$_{2.5}$-bound trace elements at the coastal area in southwestern Taiwan. The results revealed that both PM$_{2.5}$ levels and the contribution of PM$_{2.5}$-bound trace elements in cold season were higher than those in hot season. The results agreed with those of previous research, indicating that seasonal variations in the PM$_{2.5}$ concentration and its chemical constituents in Taiwan (Hwang et al. 2016; Tseng et al. 2016; Hsu et al. 2016). The effects of synoptic weather patterns, local meteorological conditions, and topography over the region of southwestern Taiwan could be explained by the seasonal variations on PM$_{2.5}$ mass concentrations and contribution of PM$_{2.5}$-bound trace elements in the current study. Taiwan, located in the East Asian subtropical region and with the Central Mountain Range (CMR) running from the north of the island to the south, is predominantly influenced by the northeasterly monsoon from late fall to spring and by the southwesterly monsoon from summer to mid-fall. In cold season, the land-sea breeze flows and mountain valley winds occur frequently formed under weak synoptic scale forcing which could enhance the accumulation of air pollutants in southwestern Taiwan (Cheng et al. 2013). Furthermore, during the cold season, the southward Asian high-pressure system frequently transports atmospheric PM from the Asian continent to Taiwan, resulting in the increase of PM over the coastline in southern Taiwan. On the other hand, during the hot season, the mesoscale convective system along with the strong southwesterly monsoonal flow can bring intermittent precipitation to southern Taiwan. Abundant precipitation could remove the primary PM$_{2.5}$ and secondary

precursors (such as SO$_2$ and NO$_x$) which reduce the mass concentrations of PM$_{2.5}$ and PM$_{2.5}$-bound trace elements.

WS was negatively associated with PM$_{2.5}$ mass concentrations in the present study, similar to the findings of other studies in Taiwan (Tsai et al. 2016), China (Zhang et al. 2015; Guo et al. 2017), Japan (Wang and Ogawa 2015), and Europe (Vardoulakis and Kassomenos 2008; Hussein et al. 2005). The stagnant weather with weak wind and relatively low boundary layer height lead to the favorable atmosphere conditions for accumulation, formation, and processing of aerosols (Chen et al. 2008). However, a study conducted by Wang and Ogawa (2015) indicated that there was a threshold in the correlations between WS and PM$_{2.5}$, negative correlation between PM$_{2.5}$ and WS lower than 3 m/s, and positive correlation between PM$_{2.5}$ and WS higher than 3 m/s in Nagasaki, Japan. Higher WS may enhance the PM$_{2.5}$ levels by transporting large quantities of PM$_{2.5}$ from far away and producing the fugitive dust. On the other hand, wind direction might play an important role in influencing seasonal variability on atmospheric PM$_{2.5}$ levels and its trace elements. Weak wind speeds usually occur on the lee side of the CMR over southwest Taiwan due to topographical blocking. Chiayi County is situated on the lee side of the CMR and exhibits low WS with an average value of 1.54 m s^{-1} (in cold season) and 1.51 m s^{-1} (in hot season). Further studies are needed and will be helpful to clarify the influence of meteorological conditions (e.g., temperature, RH, and wind activity) on PM.

The EF value of trace elements could be employed for source identification (crustal or anthropogenic) and possible pollution sources of the trace element abundances for atmospheric PM (Li et al. 2016a; Zhang et al. 2016). In this study, the major trace elements were anthropogenic origins (such as

Fig. 3 Profiles of four sources identified from the PMF model for PM$_{2.5}$ for annual
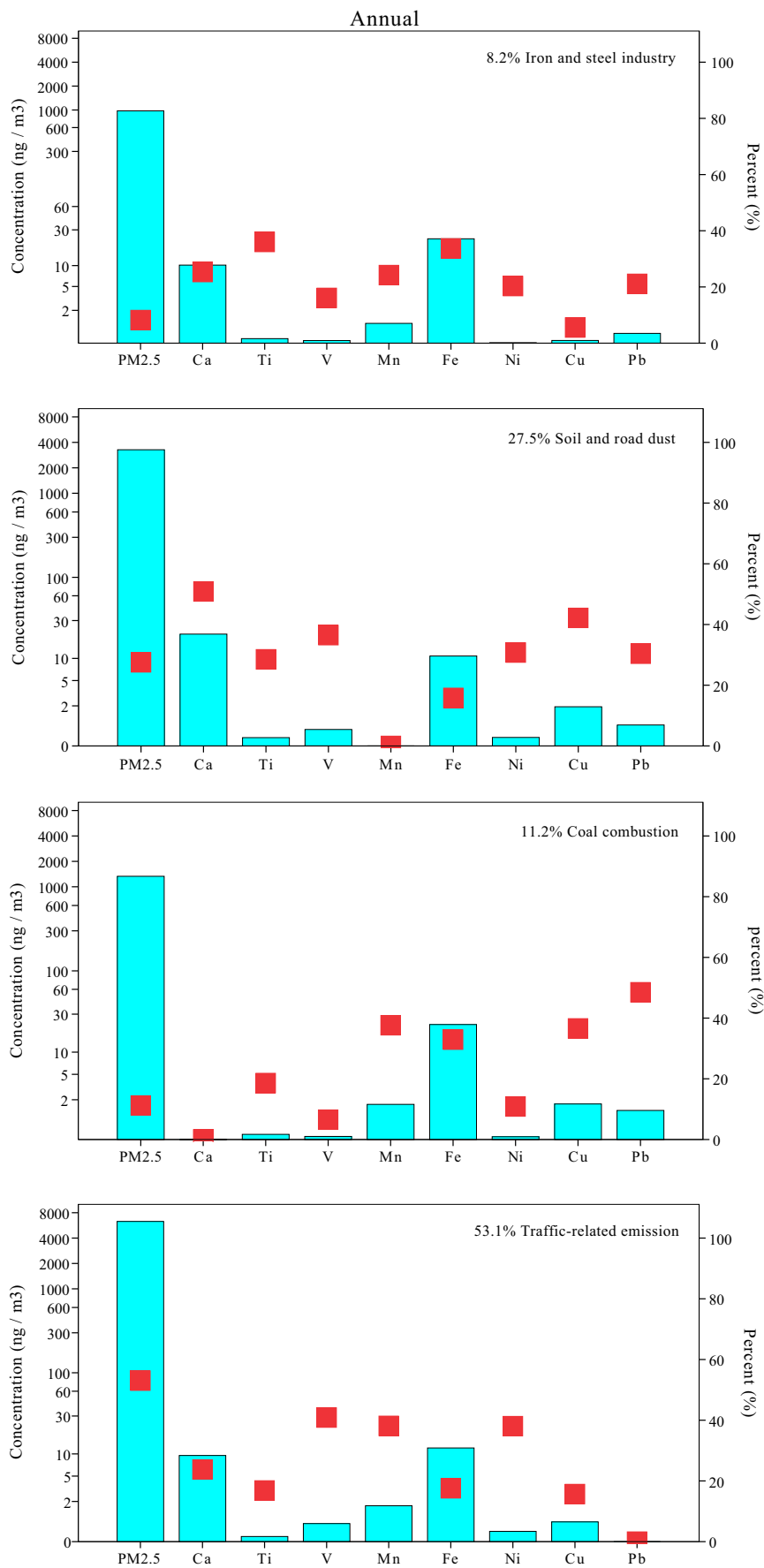
Fig. 4 Profiles of four sources identified from the PMF model for PM$_{2.5}$ for cold season
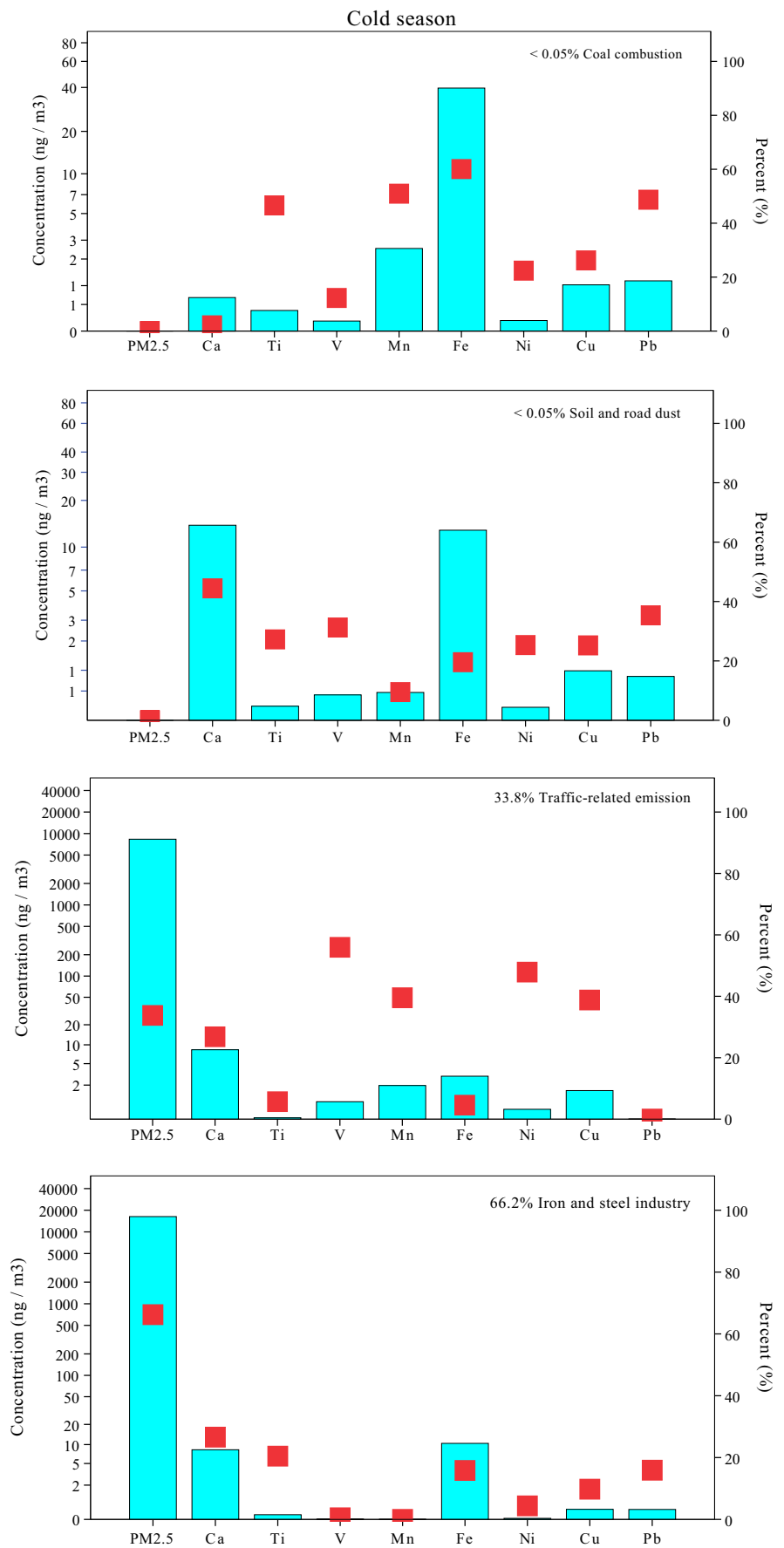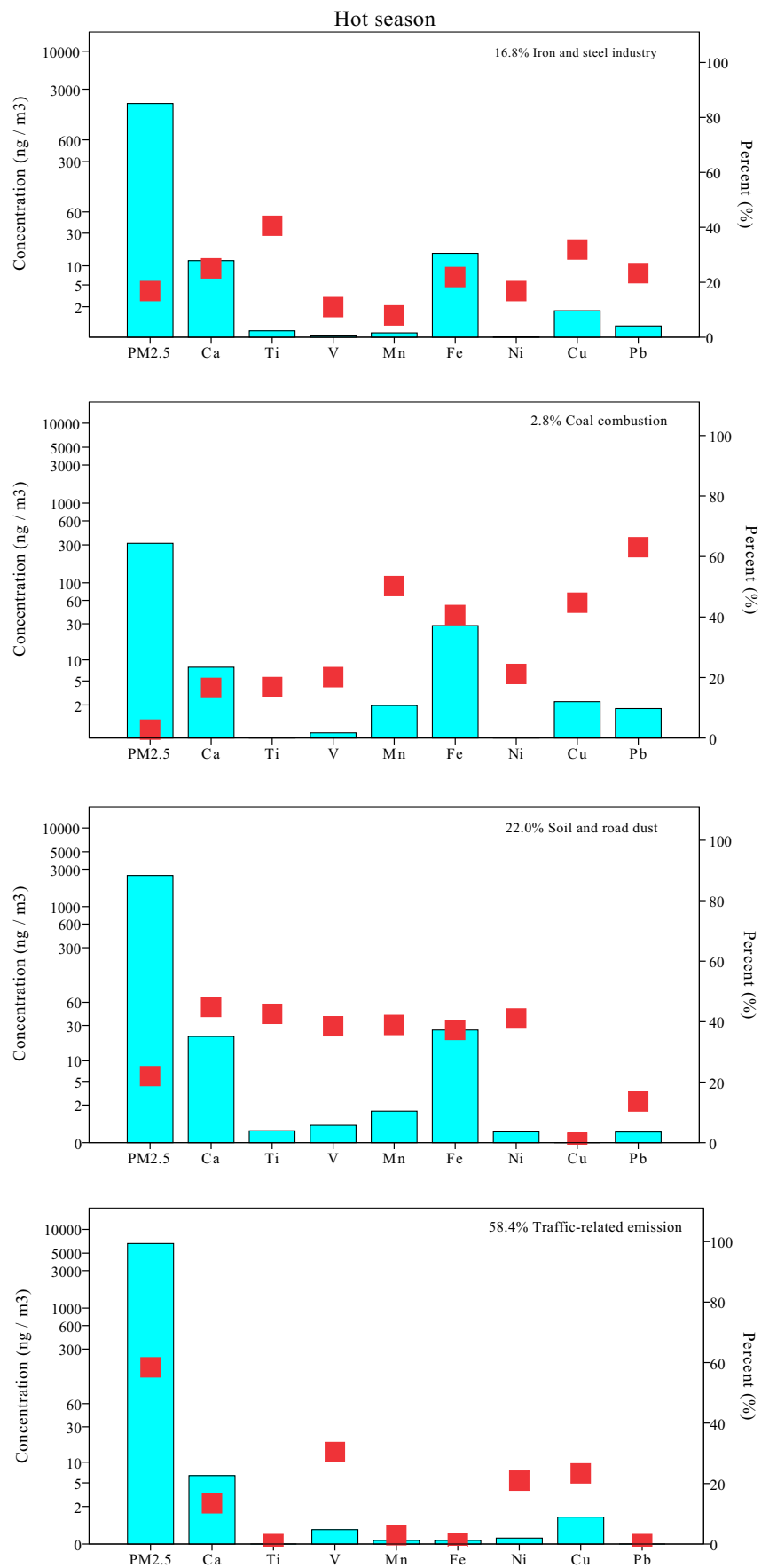
Fig. 5 Profiles of four sources identified from the PMF model for PM$_{2.5}$ for hot season

industrial boilers, petroleum plants, and steel plants) in both cold and hot seasons. The seasonal variation on the source origin of trace elements of PM$_{2.5}$ was observed. Mn and Li originated mainly from anthropogenic origins only in cold season. Similarly, Chen et al. (2015), Zhang et al. (2016), and Hsu et al. (2016) also observed seasonal variations of trace elements in PM$_{2.5}$ in Taiwan. The seasonal variations on the source origin of trace elements in PM$_{2.5}$ in the current study might be due to the local emissions, regional transported contributions (e.g., from nearby Yunlin and Changhua Counties), and long-range transport (LRT). In cold season, the prevailing northeastern monsoon winds could encourage the LRT with frontal pollutant from the Asian continent to Taiwan, which can bring abundant trace elements from anthropogenic emissions into the study area and could be explained by the seasonal variations on the source origin of trace elements in PM$_{2.5}$. Furthermore, the northeast winter monsoon with particulate pollutants passes over the heavy-polluting industrial plants, such as crude oil refining plants and coal-fired power plant, which intensively located in northern and central Taiwan, and may bring regional abundant trace elements through the atmosphere into the study area. Recently, epidemiological studies in southwestern Taiwan (Hwang et al. 2016; Hwang et al. 2017a; Hwang et al. 2017b) have reported that exposure to PM$_{2.5}$ has been associated with increased hospital admissions and emergency room visits for respiratory diseases, particularly during cold season. To sum up, the observed season variations in PM$_{2.5}$ levels and its trace elemental constituents in the current study could possibly explain the seasonal differences on the adverse effects of PM$_{2.5}$ on respiratory morbidity in southwestern Taiwan.

Through the PMF model, four potential sources of trace elements in PM$_{2.5}$ were identified: traffic-related emission, soil and road dust, coal combustion, and iron and steel industry. Similarly, (Hsu et al. 2016) concluded that coal combustion, traffic-related emission, secondary aluminum smelter, and oil combustion were important contributors of trace elements in PM$_{2.5}$ at the western coastal area of central Taiwan. The relative dominance of each identified source varies with changing seasons. The highest contributions occurred in cold season for iron and steel industry (66.2%), in hot season for traffic-related emission (58.4%), soil and road dust (22.0%), and coal combustion (2.8%). Seasonal variation of iron and steel industry was 66.2% in cold season and 16.8% in hot season. Previous study in Taiwan (Chen et al. 2015) also discovered that iron and steel industry (accounted for 30.5%) was the main source of trace elements of PM$_{2.5}$ during the period of air quality deterioration of elevated particulate matters in winter. Besides, the contribution of coal combustion (2.8%) was higher in hot season, but lower in cold season (less than 0.05%). Similar seasonal trends have been reported in central Taiwan (Tseng et al. 2016; Hsu et al. 2016). In Beijing, Zhang

et al. (2013) founded that coal combustion accounted for 18% of PM$_{2.5}$ on an annual basis, while this source was accounted for 57% of PM$_{2.5}$ in winter. In China, coal combustion has been identified as the largest contributor to the PM$_{2.5}$; moreover, coal is the main fuel used for heating in cold weather (Zhao et al. 2013). However, domestic heating is not required during winter in Taiwan. Furthermore, according to the Taiwan Energy Statistics Handbook 2016, coal-fired power plants accounted for approximately 29.4% (in 2016) of total electricity generation in Taiwan, and there was higher coal combustion in hot season than in cold season (MOEA 2017). On the other hand, the contribution of traffic-related emission was higher in hot season (58.4%), but lower in cold season (33.8%). This observation in respect to seasons was similar to the results reported by Hsu et al. (2016) who found that contribution of traffic-related emission was higher in summer (15%), but lower in winter (4.5%) in central Taiwan. Weather conditions can affect traffic intensities and traffic demand; Cools et al. (2010) indicated that high temperature significantly increases traffic density in Belgium. In Taiwan, Lin et al. (2015) observed that every 1 °C rise in temperature was associated with 0.8% increase in the hourly incidence of traffic accidents. Yang et al. (2010) reported that in Taiwan, weather factors (such as temperature, humidity, and rainfall) affect the vehicular velocity. The seasonal variation on the contribution of traffic-related emission might be governed by the variability in emission strengths, for instance, higher traffic density in hot season than in cold season. That is possibly due to high temperature that might increase traffic intensity because of the increase in outdoor activity and the volume of traffic.

The toxicity of PM$_{2.5}$ may be influenced by PM$_{2.5}$-bound composition; epidemiological research in southwestern Taiwan indicated that PM$_{2.5}$-induced effects varied by season and with higher adverse effects in cold season (Hwang et al. 2016; Hwang et al. 2017a; Hwang et al. 2017b). In cold season, trace elements (Ca, Ti, Pb, and Fe) that emitted from iron and steel industry might enhance the toxicity of PM$_{2.5}$ in southwestern Taiwan. On the other hand, it is worth noting that traffic-related emission was the main contribution source for trace elements in PM$_{2.5}$ in both cold (Ni, Mn, and Cu) and hot seasons (Ni, V, and Cu). The results suggest that controlling the emissions from the traffic-related emission and iron and steel industry may be the important strategies to improve the air quality in southwestern Taiwan.

This study has some limitations which have to be pointed out. Due to budget constraints, we only monitored PM$_{2.5}$ at one station, and the data is of limited use. More parallel sample collecting sites are required for more accurate results and conclusions. Additionally, in the present study, multiple methods were used, including EF analysis and PMF model, to identify potential sources of PM$_{2.5}$-bound trace elements. However, these methods were bulk analytical methods and

only focused on the chemical composition of aerosol particle mass, while properties of individual particles could not be provided. Individual particle analyses, scanning electron microscopy and transmission electron microscopy, can provide detailed information on the size, compositions, morphologies, structures and mixing states of individual aerosol particles. Such information should be useful for evaluating the environmental and health effects of $PM_{2.5}$. To fully evaluate the environmental and health effects posed by $PM_{2.5}$, further studies should expand both the quantitative and qualitative uses of geochemical techniques in source apportionment studies.

Conclusions

This study concludes that atmospheric $PM_{2.5}$ mass concentration and contributions of $PM_{2.5}$-bound trace elements were varied by season at the coastal area, in Chiayi County of southwestern Taiwan. $PM_{2.5}$ mass concentration was higher in cold season than in hot season. The relative dominance of each identified source varies with changing seasons. The highest contributions occurred in cold season for iron and steel industry (66.2%), in hot season for traffic-related emission (58.4%), soil and road dust (22.0%), and coal combustion (2.8%). The effects of synoptic weather patterns, meteorological factors, and topography over the region of southwestern Taiwan could possibly be explained by the observed seasonal differences in $PM_{2.5}$ levels and contributions of $PM_{2.5}$-bound trace elements in the current study. Finally, the present results suggest that controlling the emissions from traffic-related emission and iron and steel industry may be the important strategies to improve the air quality in southwestern Taiwan.

Compliance with ethical standards This study was approved by the Institutional Review Board (IRB) of Chang Gung Medical Foundation (201601350B1).

Conflict of interest The authors declare that they have no conflict of interest.

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