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Ambient $PM_{2.5}$ in the residential area near industrial complexes: Spatiotemporal variation, source apportionment, and health impact

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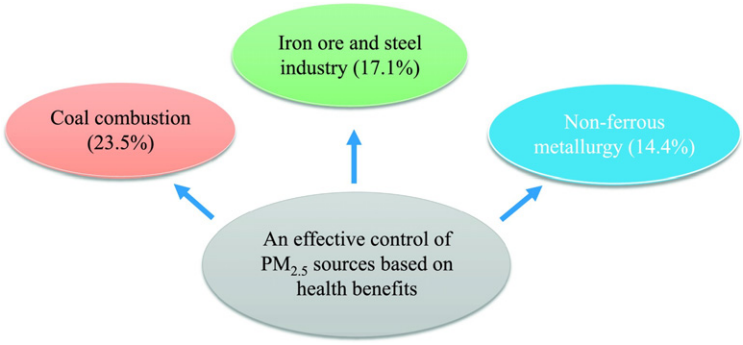
HIGHLIGHTS

- Chemical characteristics in $PM_{2.5}$ were comprehensively investigated.
- The $PM_{2.5}$ and chemical compositions were higher in winter than other seasons.
- Seven $PM_{2.5}$ sources with distinctive tracers were identified.
- Respiratory physician visits attributed to $PM_{2.5}$ for elders were estimated.
- The control strategy of sources as considering health benefits was proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

This study systemically investigated the ambient $PM_{2.5}$ (n = 108) with comprehensive analyses of the chemical composition, identification of the potential contributors, and estimation of the resultant respiratory physician visits in the residential regions near energy-consuming and high-polluting industries in central Taiwan. The positive matrix fraction (PMF) model with chemical profiles of trace metals, water-soluble ions, and organic/elemental carbons (OC/EC) was applied to quantify the potential sources of $PM_{2.5}$. The influences of local sources were also explored using the conditional probability function (CPF). Associations between the daily $PM_{2.5}$ concentration and the risk of respiratory physician visits for the elderly (≥65 years of age) were estimated using time-series analysis. A seasonal variation, with higher concentrations of $PM_{2.5}$, metals (As, Cd, Sb, and Pb), OC/EC and ions (i.e., NO^{3-}, SO_4^{2-} and NH_4^+) in the winter than in the spring and summer, was observed. Overall, an increase of 10 μg m^{-3} in the same-day $PM_{2.5}$ was associated with an ~2% (95% CI: 1.5%–2.5%) increase in respiratory physician visits. Considering the health benefits of an effective reduction, we suggest that the emission from coal combustion (23.5%), iron ore and

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steel industry (17.1%), and non-ferrous metallurgy (14.4%), accounting for ~70% of the primary $PM_{2.5}$ in the winter are prioritized to control.

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1. Introduction

Fine particles ($PM_{2.5}$, particulate matter (PM) ≦ 2.5 μm in aerodynamic diameter) are a complex mixture with various shapes, sizes, and chemical components (such as sulfate, nitrate, ammonium, inorganic and organic carbons, and trace elements). $PM_{2.5}$ can affect the atmospheric visibility, play key roles in the formation of acid rain and climate change, and deteriorate the local and regional air quality. Exposure to ambient $PM_{2.5}$ has been recognized as one of the leading causes of adverse health outcomes in relation to cardiopulmonary morbidity and mortality (Cascio et al., 2009; Dockery et al., 1993; Dominici et al., 2006; Pope et al., 2002). In addition, outdoor PM has been classified by the International Agency for Research on Cancer (IARC) as carcinogenic to humans (Group 1). Given these reasons, the governments in various countries have enforced strict air quality standards for $PM_{2.5}$.

Although Taiwan has a regulatory history in terms of its ongoing efforts to protect public health from ambient particle pollutants, overall the $PM_{2.5}$ level (annual mean = ~25 μg m^{-3} in 2014) still exceeds the guideline limit set by the Taiwan EPA (15 μg m^{-3}). In particular, the residential regions (such as Changhua and Yunlin Counties) near energy-consuming and high-polluting industries in central Taiwan, have a poor air quality of $PM_{2.5}$ (annual mean = 30 μg m^{-3}) that is usually attributed to their emissions but rarely to be clarified. Within the region, frequently occurring episodes of elevated PMs during the winter period caused by both local emissions with a poor dispersion conditions and regional contributions from seasonal monsoons have been reported (Chen et al., 2015; Kuo et al., 2010; Kuo et al., 2013; Lin et al., 2004). As a result, a number of protests against the poor air quality of $PM_{2.5}$ have been launched by residents and environmentalists who claim that their inhaled $PM_{2.5}$ is predominantly from surrounding industrial emissions which is likely to induce adverse health effects. In response to public concerns about environmental health, numerous investigations on particulate air pollutants and associated metals/PAHs have been conducted in this disputed area (Chen et al., 2015; 2016; Hsu et al., 2016; Kuo et al., 2013; Liao et al., 2015). However, significant gaps to be filled still exit, such as the lack of systematically comprehensive investigations on employing $PM_{2.5}$ chemical profiles with spatiotemporal variations and the relevant source apportionment.

The receptor-based source apportionment of PM, which can identify source categories and quantify source contributions, has been widely performed worldwide (Belis et al., 2013; Viana et al., 2008). One technique, positive matrix factorization (PMF) based on the contents of ionic components, carbons and trace metals in particles has been increasingly applied in many studies (Contini et al., 2014; Stortini et al., 2009; Tao et al., 2014) due to its advantages over other receptor models (Liang et al., 2016). It is recommended that the conditional probability function (CPF) and potential source contribution function (PSCF) can be incorporated with PMF results to qualify the contribution of each identified local and long-range transport source in a more accurate way (Heo et al., 2009; Kang et al., 2006; Kim et al., 2003; Lee and Hopke, 2006).

In addition, the associated outcomes/diseases (such as cardiopulmonary effects) on residents attributed to ambient $PM_{2.5}$ and the resultant sources are not clear within this area, which is important for the development of control measures directly based on the health burden. Beyond the particle mass metric, many studies have also indicated that the toxicity responses or adverse health outcomes are related to the chemical constituents of $PM_{2.5}$, which can be referred to specific emission sources (Bell et al., 2010; Chen and Lippmann, 2009; Franklin et al., 2008; Gehring et al., 2015). For instance, Bell et al. (2014) indicated that the risk of cardiovascular hospitalization is associated with $PM_{2.5}$ calcium, black carbon, vanadium, and zinc, which could be further referred to the contribution of $PM_{2.5}$ road dust. Thus, to develop more effective control strategies, the investigation of the $PM_{2.5}$ source apportionment linking to the health effects is crucial. Given the described research gaps, a mission-oriented project of $PM_{2.5}$ measurements and health impact analyses in Changhua and Yunlin Counties was conducted by the National Health Research Institutes in Taiwan. This study aimed to investigate the ambient $PM_{2.5}$ with comprehensive analyses of its chemical composition, identify its potential contributors, and evaluate resultant respiratory physician visits. The assessment of the health impacts enables the estimations of both the burden of disease attributable to air pollution and the potential benefit from policies driven to improve the air quality (Boldo et al., 2006; Kunzli et al., 2000). This study also sought to propose a $PM_{2.5}$ control measure from sources in accordance with the abatement of the health burden.

2. Materials and methods

2.1. Sampling sites and $PM_{2.5}$ collection

The sampling was conducted in residential areas of Changhua (23° 53′ N, 120° 23′ E, 16 m above sea level) and Yunlin Counties (23° 42′ N, 120° 22′ E, 8 m above sea level) in central Taiwan. The selected sampling sites are within approximately 20 km radius of the Mailiao petrochemical complex (which is the largest oil refinery in Taiwan and the largest naphtha cracking plant in the world) containing a coal-fired power plant (ranked No. 8 worldwide in terms of carbon dioxide emissions). The sites are also 20 km from the Chang-Bin industrial park (a cluster of factories engaging in non-ferrous metal smelting, tire production, steel manufacturing, semiconductor manufacturing, and glass production) and within 80 km of the Taichung coal-fired power plant (ranked No.1 worldwide in terms of carbon dioxide emissions) and integrated iron ore and steel manufacturing (Fig. 1). Several provincial routes and a highway across or near the study area are observed (Fig. 1). Approximately 1,291,000 and 705,400 people live in Changhua and Yunlin Counties, respectively.

Due to similar patterns of past (5-year, 2008–2012) $PM_{2.5}$ levels and meteorological conditions (such as temperature, humidity, wind speed and wind direction) being obtained from the Taiwan EPA data for the fall and winter, we conducted air sampling in the spring, summer and winter to determine the average concentrations with annual and seasonal variations. In Fig. S1, three main prevailing wind directions from the NE, W and S for the winter, summer and spring, respectively, could be observed in the study area. We chose six sampling sites (sites A–F in Fig. 1) at two distances (<5 and 10–20 km) from the Mailiao petrochemical complex because it has been deemed a prime source of the poor air quality and adverse health effects. The site selections were based on the three prevailing wind directions within a year to distinguish the spatial variations in the $PM_{2.5}$. Sites A and B together were defined as the north site; Sites C and D together were defined as the east site; and Sites E and F together were defined as the south site. A total of 108 daily samples with filter-based $PM_{2.5}$ were collected on the roof (9 m height) of elementary schools at these six sites in the spring (from 5th May) and summer (from 4th August) of 2014 and the winter (starting on 26th January) of 2015. Two high-volume samplers (BGI PQ200) were used at each site to synchronously collect $PM_{2.5}$ samples on both PTFE and quartz filters with a diameter of 47 mm at a flow

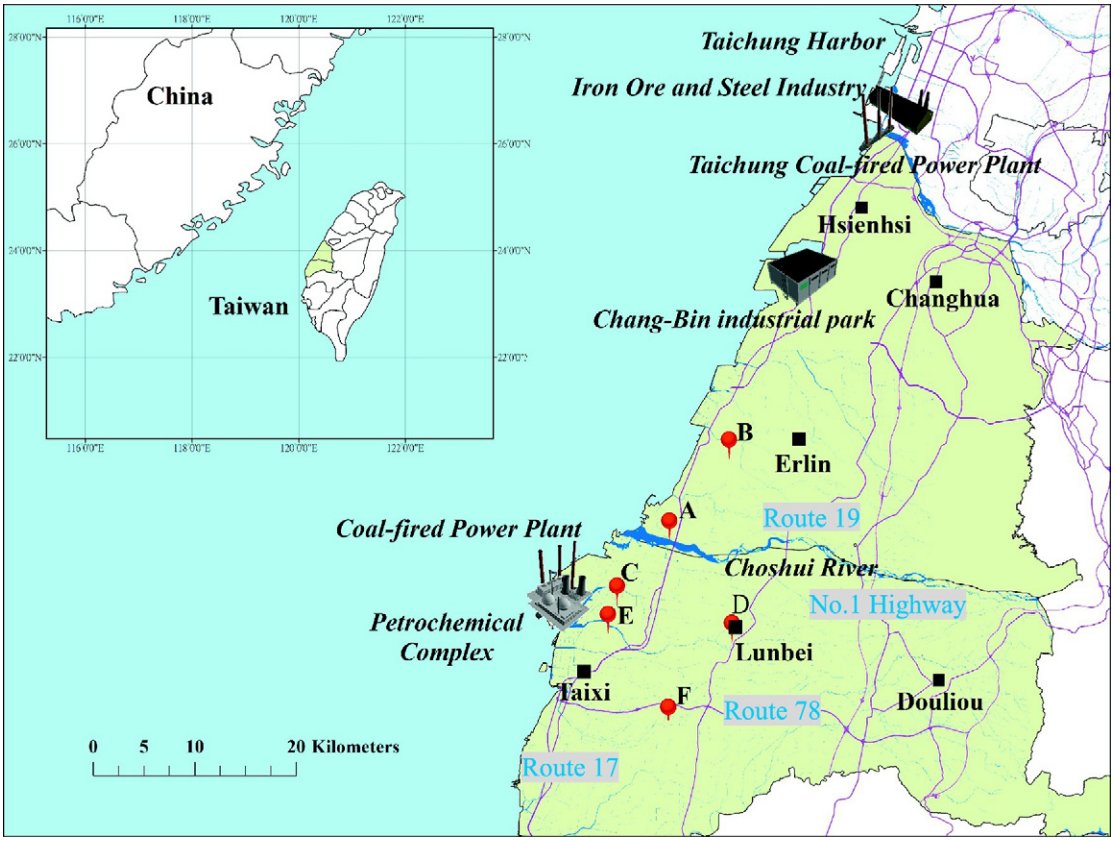

Fig. 1. Overview of the sampling sites (A–F) and monitoring stations (■) of the Taiwan EPA for Changhua and Yunlin Counties.

rate of 16.7 L min^{-1} for 24 h. We pre-baked the quartz filters at 550 °C for 6 h to eliminate their carbon blanks and we recorded the total volume of air that passed through a filter for each sample. Using a calibrator rotameter (MesaLabs Defender 520), we readjusted the sampling flow rate for each campaign. The $PM_{2.5}$ mass was determined by weighing the PTFE filter (and weighing the quartz filter for reference) using an electronic microbalance (Mettler-Toledo MX5). Before and after sampling, the filters were equilibrated in temperature/relative humidity controlled conditions (23 ± 1 °C and 40 ± 5% relative humidity) for 24 h.

2.2. Chemical analysis

Half of the quartz filters were extracted with 10 mL of ultrapure water in an ultrasonic bath for 60 min to analyze the water-soluble ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, NO_3^-, SO_4^{2-} and Cl^-). The water extracts were filtered through a 0.22 μm pore-size 33-mm filter (MIL, SLGS033) and then stored at 4 °C for later analysis. The ions were determined in a Shimadzu system (HPLC, Shimadzu) consisting of an LC-10Ai pump, a CDD-10Avp conductometric detector (0.25 μL flow cell) and a CTO-20AC column oven. The anions were analyzed by a system equipped with a guard column (IonPac™ AG12A, 4 × 50 mm), analytical column (AS12A, 4 × 200 mm) and anion self-regenerating suppressor (ASRS, 300/4 mm) with 0.0017 mM $NaHCO_3$ and 0.0018 mM Na_2CO_3 as eluents. For the cation analysis, the system was equipped with a guard column (IonPac™ CG12A, 4 × 50 mm), analytical column (CS12A, 4 × 250 mm) and cation self-regenerating suppressor (CERS, 500/4 mm) using 20.0 mM MSA (methane sulfonic acid) as an eluent. The detection limits of the water-soluble ions were 0.1, 0.1, 0.1, 0.2, and 0.2 μg mL^{-1} for Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}, respectively, and 0.1 μg mL^{-1} for NO_3^-, SO_4^{2-},and Cl^-. The other half of the quartz filters were used to determine the total carbon (TC) and elemental carbon (EC) using an elemental analyzer (Heraeus Elemental Analyzer CHN-O-Rapid). More details can be found in a previous study (Tsai and Chen, 2006).

Half of the PTFE filters were used to analyze for trace elements, including Mg, Al, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Sr, Mo, Cd, Sb, Ba, Pb and 7 rare earth elements (REEs, La, Ce, Pr, Nd, Sm, Yb and Lu) using a microwave digestion system (Anton-Paar Multiwave 3000) and inductively coupled plasma mass spectrometry (ICP-MS, AGILENT-7700×). More details can be referred to our previous study (Hsu et al., 2016). The other half of the PTFE filters were used to analyze for polycyclic aromatic hydrocarbons (PAHs) but the data are not shown here.

2.3. Positive matrix factorization

Positive matrix factorization (PMF) was used to determine the source contributions to the $PM_{2.5}$ in our study area. The details of PMF can be found in our previous study (Hsu et al., 2016) and a number of references (Han et al., 2006; Lee and Hopke, 2006; Song et al., 2006). In this study, we employed PMF 5.0 and input all analyzed species of $PM_{2.5}$ into the model computation.

2.4. Conditional probability function (CPF)

We performed the CPF based on the daily source contributions resulting from the PMF, coupled with the surface wind direction values to estimate the influence of local sources from various wind direction (Kim et al., 2003). The CPF is defined as

$$\mathrm{CPF} = \frac{m_{\Delta\theta}}{n_{\Delta\theta}} \quad (1)$$

where $m_{\Delta\theta}$ is the number of occurrences from wind sector $\Delta\theta$ that exceeded the threshold criterion, and $n_{\Delta\theta}$ is the total number of data

from that wind sector. Here, $\Delta\theta$ was set at 22.5°, and calm winds (<1 m s^{-1}) were excluded from this analysis. The threshold was set at ≥ the 20th percentile value of the fractional source contributions for each source because these sources are likely to be located in directions that have high probability values. Although the aerosols from long-range transported source locations could be estimated by using a PSCF model, the estimation of the local source contributions is of interest to us for control.

2.5. Estimation of $PM_{2.5}$-related health impact

The relative risk (RR) and attributable number of cases with the increment in $PM_{2.5}$ concentrations among residents living in Changhua and Yunlin Counties were then calculated during the period of time. Here, the daily $PM_{2.5}$ data from 2006 to 2011 was obtained from air quality monitoring stations of the Taiwan EPA located in six townships of Changhua and Yunlin Counties. The health data on respiratory physician visits for the elderly (2101 beneficiaries with 16,537 counts) of ≥65 years of age who resided in these townships and were enrolled in the Taiwan National Health Insurance Research Database (NHIDR) during 2006–2011 were obtained. The respiratory physician visit was determined by the principal discharge diagnosis code according to the International Classification of Diseases, Ninth Revision, Clinical Modification (ICD-9-CM; codes 460–519). A township-specific season-stratified time-series analysis using the generalized additive model (GAM) with a Poisson distribution was performed as following:

$$\ln\left(\mathrm{E}\left[Y^c_{t,s}\right]\right) = \text{Intercept} + \beta x^c_{t,s} + ns\left(T^c_{t,s}, df_T\right) + \alpha^c DOW^t + ns\left(y, df_y\right) \quad (2)$$

where, $Y^c_{t,s}$ is the physician visits in township c on day t of season s; β is the coefficient relating $PM_{2.5}$ to physician visits for an increment of 10 μg m^{-3}; $x^c_{t,s}$ is the $PM_{2.5}$ level in township c on day t of season s; $ns(T_{t,s}{}^c, df_T)$ is the natural cubic spline of the temperature in township c on day t with $df_T = 5°$ of freedom; α^c is the regression coefficient relating the day of the week to physician visits in the township c; DOW^t is the day of the week on day t; and $ns(y, df_y)$ is the spline of the year (y) with $df_t = 5°$ of freedom. We only considered the single-day lag of exposure on the same day as a physician visit (lag 0). Random effect meta-analysis was used to estimate the RR for the six townships annually.

The $PM_{2.5}$-related respiratory physician visits was also calculated as following:

$$P_0 = P_E/\{1 + [(RR-1)(E-B)/10]\} \quad (3)$$

$$D_{10} = P_0 \times (RR-1) \quad (4)$$

where, P_E is the observed or current frequency of respiratory diseases; P_0 is the expected frequency of respiratory diseases at the reference level; E is the observed or current exposure level; B is the reference exposure level (10 μg m^{-3}). RR is the relative risk of respiratory physician visits per 10 μg m^{-3} increment; and D_{10} is the attributable number of cases per 10 μg m^{-3} increment in $PM_{2.5}$ levels. The health burden of respiratory diseases (D_{10}) treated as a mitigation scenario in this study was then applied to initiate the benefit of $PM_{2.5}$ source control.

3. Results and discussion

3.1. $PM_{2.5}$ mass concentrations and chemical compositions

Table 1 shows a statistical description of the annual and seasonal concentrations for $PM_{2.5}$ mass, and associated OC, EC, water-soluble ions and metals obtained from the selected sampling sites. Based on the coefficient of divergence (CD) results (ranging from 0.13 to 0.26; see Fig. S2 in the Supplementary material and the methods), the low spatial heterogeneity among sampling sites (A–F) for the concentrations of $PM_{2.5}$ mass and chemical compositions allows us to integrate those data for further analysis. Here, the annual mean concentration of the $PM_{2.5}$ mass was 24.1 ± 12.8 μg m^{-3}, which exceeded the annual air quality standard of $PM_{2.5}$ (15 μg m^{-3}) set by the Taiwan EPA. Higher $PM_{2.5}$ concentrations were obtained in the winter (36.1 ± 12.9 μg m^{-3}) than in the spring (20.8 ± 11.2 μg m^{-3}) or summer (18.0 ± 6.6 μg m^{-3}), likely due to local emissions with poor dispersion caused by the low mixing height and/or the long-range transport of air pollutants from continental China coming along with the winter monsoons. In addition, the local crustal materials (including river and road dust) resuspended into the ambient air through stronger winds in the winter may contribute, in part, to the higher $PM_{2.5}$ concentrations. In addition to the seasonal variation, the daily variation in $PM_{2.5}$ concentrations affected by meteorological factors has also been clarified. A previous study showed that the prior day wind speed, precipitation, and sunlight hours were negatively correlated with the current $PM_{2.5}$ concentration, while the air pressure 3 days earlier had a significant positive correlation (Huang et al., 2015). However, there is no uniform conclusion on the issue of temperature. While the previous studies indicated a positive correlation of temperature with daily $PM_{2.5}$ concentrations (Wang and Ogawa, 2015; Tai et al., 2010), Huang et al. (2015) presented the insignificant result.

For the annual trace metals, in addition to Al and Fe (10 to 1000 ng m^{-3}), the most abundance were Ti, Cr and Pb (10 to 100 ng m^{-3}), followed by V, Mn, Ni, Cu, Zn, As, Se, Sr, Mo and Ba(1 to 10 ng m^{-3}). The Lanthanides (La, Ce, Pr, Nd, Sm, Yb, Lu), Co, Cd, Sb,

Table 1
Concentrations of $PM_{2.5}$ (μg m^{-3}) and associated OC (μg m^{-3}), EC (μg m^{-3}), ions (μg m^{-3}) and metals (ng m^{-3}) including in all sites for annual and seasonal periods.

Species	Spring (n = 42)		Summer (n = 36)		Winter (n = 30)		Annual (n = 108)	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
$PM_{2.5}$	20.8	11.2	18.0	6.6	36.1	12.9	24.1	12.8
EC	1.55	0.82	0.827	0.449	2.41	1.09	1.55	1.01
OC	3.53	1.95	2.92	1.53	5.50	2.52	3.87	2.24
Na^+	0.309	0.192	0.461	0.273	0.793	0.231	0.494	0.303
Mg^{2+}	0.123	0.025	0.130	0.018	0.594	0.039	0.256	0.212
K^+	0.264	0.108	0.253	0.097	0.406	0.127	0.300	0.127
Ca^{2+}	0.436	0.528	0.286	0.072	0.948	0.246	0.528	0.444
Cl^-	0.428	0.447	0.374	0.523	0.964	0.603	0.559	0.573
NO^{3-}	1.94	2.04	1.20	1.30	4.82	2.53	2.49	2.47
SO_4^{2-}	2.38	1.57	2.14	1.10	4.22	2.10	2.81	1.82
NH_4^+	2.09	1.86	1.53	1.15	4.54	2.55	2.59	2.25
Al	853	185	904	110	1000	337	911	227
Ti	14.7	6.35	42.8	179	25.0	24.9	27.0	104
V	5.75	4.22	5.92	4.25	5.34	3.03	5.69	3.91
Cr	34.1	8.0	29.3	19.4	26.1	3.0	30.3	12.7
Mn	5.92	3.61	4.56	1.93	14.5	8.01	7.86	6.42
Fe	97.2	53.8	115	117	278	219	153	157
Co	0.234	0.411	0.164	0.118	0.233	0.145	0.210	0.275
Ni	6.17	12.1	3.28	5.20	4.17	1.55	4.65	8.20
Cu	4.86	3.30	5.11	3.38	9.19	6.32	6.15	4.73
Zn	49.7	38.0	34.6	12.9	109	44	61.2	45.4
As	1.51	1.02	1.20	0.56	2.60	1.85	1.71	1.32
Se	8.22	7.93	3.85	4.36	1.19	0.79	4.81	6.25
Sr	1.26	0.79	2.81	9.67	1.92	1.06	1.96	5.62
Mo	3.13	4.19	1.07	1.21	1.04	1.01	1.86	2.92
Cd	0.233	0.256	0.159	0.107	0.713	0.671	0.342	0.453
Sb	0.867	0.588	0.519	0.265	1.13	0.809	0.822	0.625
Ba	6.99	12.31	11.9	40.0	4.69	3.83	7.98	24.39
Pb	9.49	9.22	3.71	2.12	20.6	14.1	10.7	11.5
Pt	0.009	0.004	0.009	0.012	0.013	0.002	0.010	0.007
Ce	0.159	0.105	0.129	0.138	0.330	0.343	0.197	0.222
La	0.107	0.060	0.104	0.132	0.277	0.215	0.154	0.160
Nd	0.059	0.055	0.042	0.019	0.107	0.150	0.066	0.090
Pr	0.017	0.014	0.009	0.005	0.033	0.036	0.019	0.023
Sm	0.031	0.020	0.021	0.013	0.045	0.038	0.032	0.026
Yb	0.007	0.005	0.004	0.002	0.009	0.008	0.007	0.005
Lu	0.004	0.003	0.002	0.001	0.003	0.001	0.003	0.002

and Pt were at even lower concentrations (<1 ng m^{-3}). The WHO (2000) published guidelines for Air Quality in 2000 for certain major heavy metals such as Pb, V, As, Mn, Ni, Cd and Cr(VI), and our data on the annual mean concentrations for As, Ni and adjusted Cr(VI) (Hsu et al., 2016) in $PM_{2.5}$ exceeded the guideline limits (which corresponding to an excess lifetime cancer risk of 10^{-6} for As, Ni and Cr(VI)). Seasonal variation in the trace metals, with the highest levels for Mn, Fe, Cu, Zn, As, Cd, Sb, Pb, Pt, and lanthanides in the winter and for Ti, Sr, and Ba in the summer, were also observed. We adopted enrichment factor (EF) analysis to roughly delineate the crustal and anthropogenic sources for $PM_{2.5}$ trace metals. The detailed method could be found in our previous works (Chen et al., 2015; Hsu et al., 2016). Fig. S3 shows that the EF levels of selected trace metals varied similarly across all seasons. In each season, Cr, Cu, Zn, As, Se, Mo, Cd, Sb and Pb with higher EF values (≥5) were predominantly from anthropogenic emissions while Ti, Mn, Fe, Sr, Ba, and the lanthanides (La, Ce, Pr, Nd, Sm, Yb, Lu) with lower EF values (<1) were customarily of crustal origin. V and Co, with EFs of 1.0–5.0, were considered to be from both anthropogenic and crustal sources. It seems that the high concentrations of Ti, Sr, and Ba in the summer described before is associated with crustal/soil dusts with the prevailing wind directions of W, SW and S from the Chianan plain (containing the largest agriculture field in Taiwan).

Of the water-soluble ions, SO_4^{2-} had the highest concentration with an annual mean of 2.81 ± 1.82 μg m^{-3}, followed by NH_4^+ (2.59 ± 2.25 μg m^{-3}), NO_3^- (2.49 ± 2.47 μg m^{-3}), Cl^- (0.56 ± 0.57 μg m^{-3}), Ca^{2+} (0.528 ± 0.444 μg m^{-3}), Na^+ (0.494 ± 0.303 μg m^{-3}), K^+ (0.300 ± 0.127 μg m^{-3}), and Mg^{2+} (0.256 ± 0.212 μg m^{-3}). On average, the combinations of SO_4^{2-}, NO_3^-, and NH_4^+, within the secondary inorganic aerosols accounted for 78.7% of the total ionic concentration. For the seasonal variation, we found the highest level in the winter and the lowest level in the summer for all water-soluble ions. The seasonal variation of the secondary inorganic aerosols was consistent with those of SO_2 and NO_2, which are precursors of SO_4^{2-} and NO_3^- that were obtained at nearby EAP air quality monitoring sites (data not shown here).

The OC concentration was higher than that of EC for all seasons. Unsurprisingly, the winter (OC = 5.50 ± 2.52 μg m^{-3}, EC = 2.41 ± 1.09 μg m^{-3}) had the highest mean concentrations, followed by spring (OC = 3.54 ± 1.95 μg m^{-3}, EC = 1.55 ± 0.82 μg m^{-3}) and summer (OC = 2.92 ± 1.53 μg m^{-3}, EC = 0.827 ± 0.449 μg m^{-3}). The majority of the OC/EC ratios fell within the range of 2.3–3.8, with mean ratios of 2.3, 3.4 and 2.3 in the spring, summer and winter, respectively. These values were consistent with those observed in a previous study conducted in Taichung in central Taiwan (Chou et al., 2010), which suggested a relatively high concentration of secondary organic aerosols in the summer but of primary aerosols in the other seasons. Chou et al. (2010) also indicated that the elevated concentrations of secondary organic carbons in the summer were likely due to the increases in biogenic secondary organic aerosols precursors (Bencs et al., 2008) and the enhancement in the secondary organic aerosol yield.

3.2. Source apportionment

We eliminated 7 lanthanides originated from the crustal element exclusively (due to their EF values) and input 29 species of measured components for a PMF model. Seven main sources based on annual $PM_{2.5}$ data were identified as secondary inorganic aerosols (31.8%), coal combustion (22.5%), traffic-related emissions (18.1%), iron ore and steel industry (8.1%), oil combustion (7.5%), non-ferrous metallurgy (6.7%), and soil dust (5.2%). The relative contributions of each identified source to $PM_{2.5}$ on both seasonal and annual base are summarized in Table 2. The secondary inorganic aerosol source mostly dominated over the identified sources. The relative dominance of each $PM_{2.5}$ source varied by season. For instance, the seasonal trends showed higher contributions in the winter (23.5%) and summer (20.2%) than in the spring (4.5%) for the coal combustion, and the opposite result was observed for soil dust. The sources of iron ore and steel industry (17.1%) and non-ferrous metallurgy (14.4%) had the highest contribution to $PM_{2.5}$ in the winter time, and the oil combustion with the highest contribution was observed in the summer. Those seasonal variations in the $PM_{2.5}$ contributors are likely attributed to climate effects, such as temperature, humidity, wind speed and wind direction, as well as long-range transport.

Table 2
Relative contributions of each identified source to $PM_{2.5}$ on both seasonal and annual base.

Source	Spring	Summer	Winter	Annual
Secondary inorganic aerosol	34.1%	21.3%	21.2%	31.8%
Coal combustion	4.5%	20.2%	23.5%	22.5%
Traffic-related emission	22.6%	22.2%	7.3%	18.1%
Iron ore and steel industry	1.8%	2.1%	17.1%	8.1%
Oil combustion	11.6%	19.4%	7.9%	7.5%
Non-ferrous metallurgy	6.7%	5.5%	14.4%	6.7%
Soil dust	18.7%	9.3%	8.5%	5.2%

Fig. 2 shows the modeled source profiles for each identified source by analyzing the annual $PM_{2.5}$ data. We then further compared the time series of contributions for each source resulting from the PMF with the observed time series of selected chemical species to find those that could represent respective sources (Fig. 3). Finally, we identified the geographical origins of each $PM_{2.5}$ source using a CPF (Fig. 4) analysis to provide insights into source localization. The secondary inorganic aerosol is the first source to be called out because of its high contributions to Cl^- (57.8%), NO_3^- (56.4%), SO_4^{2-} (41.4%) and NH_4^+ (49.7%). The secondary inorganic aerosol is likely to be caused by the following sources: vehicle exhaust, coal combustion, biomass burning, oil burning, waste incineration, and household emission via their precursor gas-to-particle conversion. Indeed, the formation of the secondary inorganic aerosol depends on the concentrations of SO_2, NO_x and NH_3, relative humidity, temperature, OH/radiation, and nighttime chemistry via NO_3 (Heo et al., 2009), each of which exhibits seasonal variations. Given the dominance of the lower temperature and higher humidity over those in other two scenarios such as large amounts of NH_3 due to applying fertilization in agricultural fields in the spring (Kim et al., 2006) and secondary sulfate and ammonium through strong photochemical reactions in summer (Zhang et al., 2013), the intense generation of secondary nitrate particles is facilitated in the winter season in this study (see Table 1) (Heo et al., 2009; Seinfeld and Pandis, 1998). In the time series of daily concentrations (Fig. 3a), our results also showed maximal and minimal levels of secondary inorganic aerosol in the winter and summer, respectively. The temporal variations of the secondary inorganic aerosol were expectedly correlated with time series of NH_4^+ and Cl^- concentrations (R^2 = 0.91 by Pearson correlation analysis) (Fig. 3a).

The CPF plot (Fig. 4a) shows that the elevated secondary inorganic aerosols mainly coming from the NNE and NE directions (50% versus 20% for the other directions). The target emission sources in the western coastal area of central Taiwan are located in NNE and NE directions of the study area. These sources include the coal-fired power plant, non-ferrous industries, iron ore and steel industries, and the many shipping/fishing vessels near the Taichung Harbor. Notably, such pollutant sources impact not only the local air quality but frequently also the downwind rural areas (Comrie, 1994; Kumar et al., 2008; Yang et al., 2008). For example, both the secondary $PM_{2.5}$ and ozone typically form 30–200 km downwind from the precursors non-methane hydrocarbons (Cheng et al., 2001; Seinfeld and Pandis, 1998; Vukovich, 1994; Wang and Chen, 2008). This also explains why the air quality of our study area is similarly impacted by the emission sources located 30–50 km away.

Fig. 2 identifies factor 2 as coal combustion in view of the high percentage of Sb (73.1%), Cd (60.9%), Pb (60.4%), EC (45.5%), OC (39.5%) and As (38.7%). Many studies have suggested a link between coal combustion and high percentages of these components, where As and Sb are often considered as tracers for coal burning (Gu et al., 2010; Manoli et al., 2002; Mokhtara et al., 2014; Pacyna et al., 2007; Tian et al., 2010;

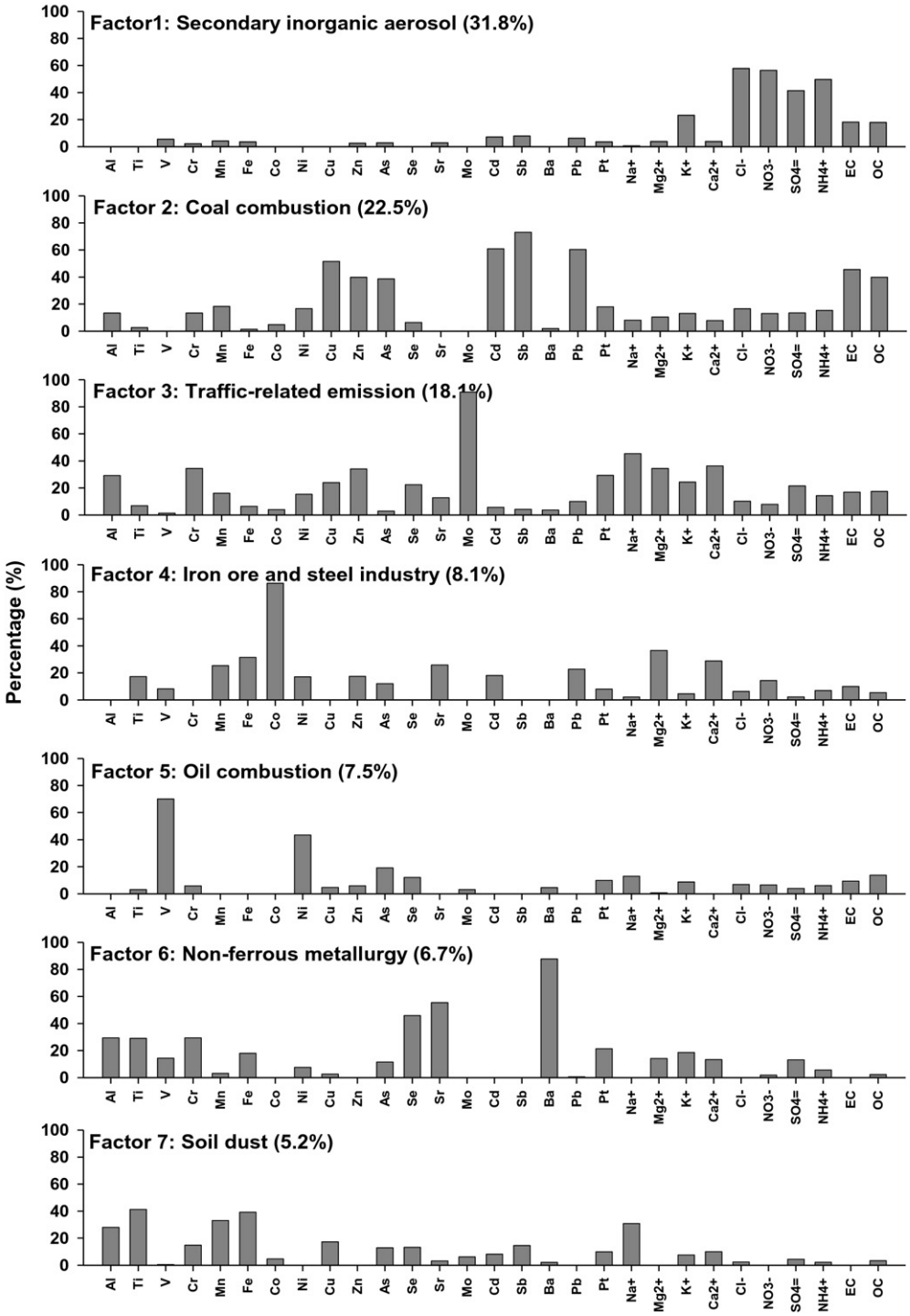

Fig. 2. Profiles of seven sources identified from the PMF model for $PM_{2.5}$.

Zhang et al., 2013). The additive of Pb to gasoline has been banned by the Taiwan government since 2000, which resulted in a decrease in vehicle-associated Pb emissions in the ambient air (Wang et al., 1998). Thus, the atmospheric $PM_{2.5}$-bound Pb in Taiwan is most likely attributable to coal combustion, not vehicle emissions. Zhang et al. (2009 and 2013) and Tian et al. (2012) have stated that the $PM_{2.5}$-bound Pb and Sb in the cities of Shanghai and Beijing in China were also related to coal combustions. Fig. 3b thus shows the strong covariation between the daily contribution of coal combustion and Sb concentration. This result also suggests that Sb, at least in Changhua and Yunlin Counties, could be an effective tracer of coal combustion. The CPF plot (Fig. 4b) shows that coal combustion, mainly from the NNE and NE and partially from the WNW, tended to influence the sampling site, which could be confirmed by exact local sources where a coal-fired power plant is in a petrochemical complex and the Taichung coal-fired power plant located at the W and NE directions, respectively. One should note that some unidentified middle- and small-scale factories using coal as fuel in those locations are also relevant.

Factor 3 represents the traffic-related emission in Fig. 2 which shows high contents of Mo (90.81%), Ca (36.3%), Cr (34.4%), Zn (34.0%) and Cu (24.2%). Previous studies have used Mo as an indicator of emission from gasoline/diesel engines (Lin et al., 2015; Wang et al., 2003). While Mo/

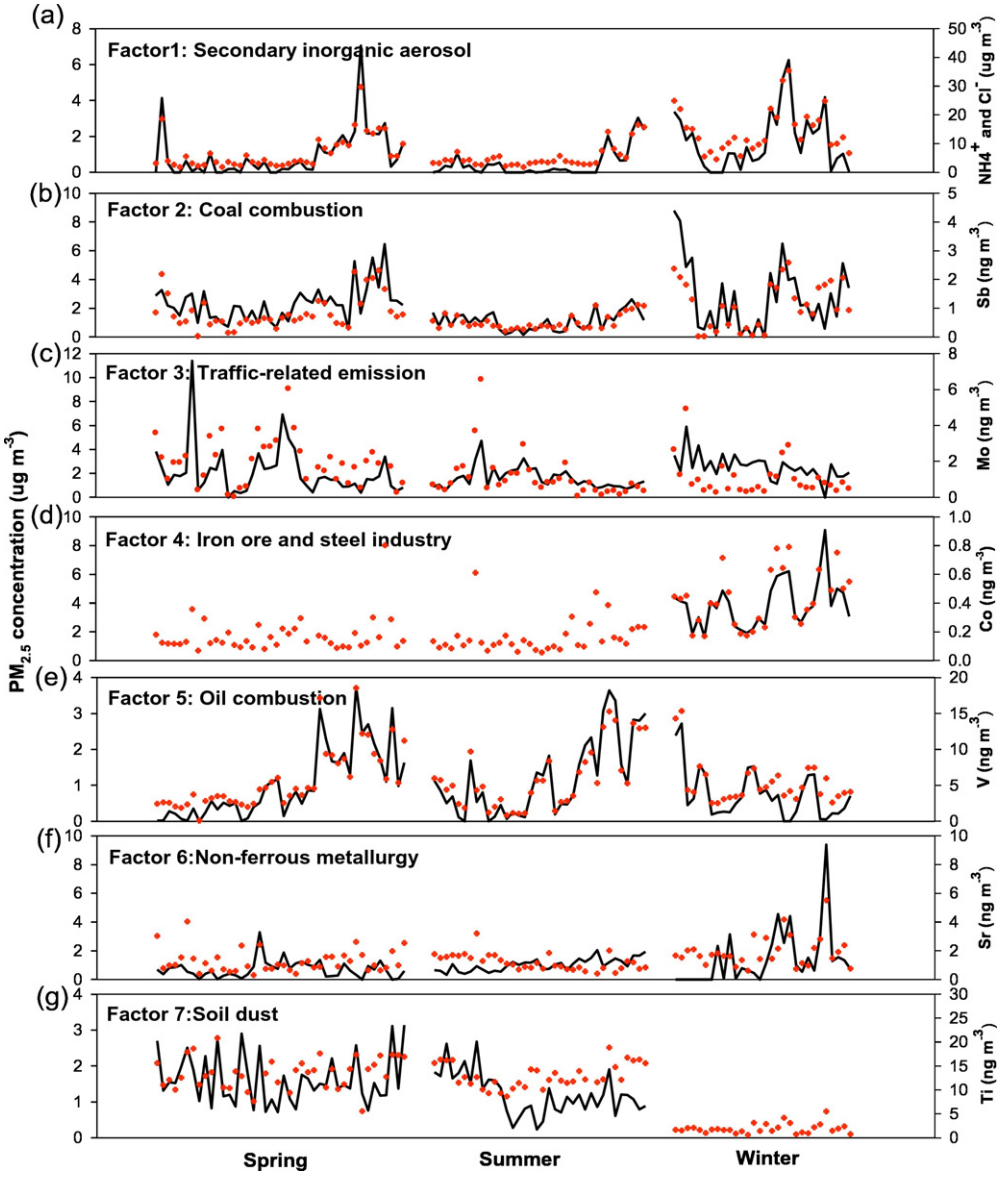

Fig. 3. Time series of daily contributions and representative tracers from each identified source during the study period.

Ni ratio less than one has usually been used to recognize the existence of traffic-related sources (Alleman et al., 2010; Mooibroek et al., 2011), our data showed Mo/Ni ratios (summer = 0.90, spring = 0.73, and winter = 0.24) less than unity. The higher loading of Ca, Cu, and Zn referring to diesel vehicles has also been reported (Lee and Hopke, 2006). The high amount of Cr and Ca were likely related to re-suspended road dust (Yongming et al., 2006). As expected, a good correlation between the time series contribution of traffic-related emissions and the daily Mo concentrations was also observed in Fig. 3c. The CPF plot (Fig. 4c) shows directions with high probabilities of the N, NNE, NE and WNW (30% on average), presenting the local origin of the traffic-related emissions. Routes 17 and 19 serve as the main commuting routes in the study area. Along these routes, vehicles and scooters, especially during rush hours, collectively emit and re-suspend the associated aerosol. Mailiao Harbor and Taichung Harbor located W (20 km radius) and NE (80 km radius) of the study area, respectively, may contribute in part to the $PM_{2.5}$ due to shipping and port activities.

Factor 4 is associated with the iron ore and steel industry based on the high contents of Co (86.2%), Fe (31.5%), Mn (25.3%) and Pb (22.8%) as shown in Fig. 2. These metals have been identified as the main components in aerosols in the iron ore and steel industry (Cheng et al., 2015; Querol et al., 2007; Tsai et al., 2007). In Fig. 3d, the temporal variation of Co is well consistent with the time series of the contributions. This result further implies that Co, at least in Changhua and Yunlin Counties, could be an effective tracer of $PM_{2.5}$ pollutants for the iron ore and steel industry. The exactly directional local source could also confirm this finding, where the results of the time series contribution (Fig. 3d) with unique peaks in the winter (prevailing wind direction from NE; Fig. S1) and high air masses from the NNE in the CPF plot (Fig. 4d) were observed while an integrated iron ore and steel manufacturing facility is located to the NE, 50 km from our study area (Fig. 1).

Factor 5 is related to oil combustion characterized by high contributions of V (64.5%) and Ni (43.1%) as shown in Fig. 2. Both V and Ni have been recognized as tracers of oil combustion for $PM_{2.5}$ (Dall'Osto et al., 2013; Lin et al., 2015; Shafer et al., 2012). An excellent correlation between the time series contribution of oil combustion and the temporal concentration for V ($R^2 = 0.92$) was also observed in Fig. 3e. In addition to a major use in oil refinery plants, heavy oil is used to supplement the power supply in industrial boilers and ships. Given that the CPF plot (Fig. 4e) shows high probabilities in the NNW and WSW for the oil combustion source, the local heavy-polluting industries (such as factories in the Chang-Bin industrial park and crude oil refining plants) and vessels

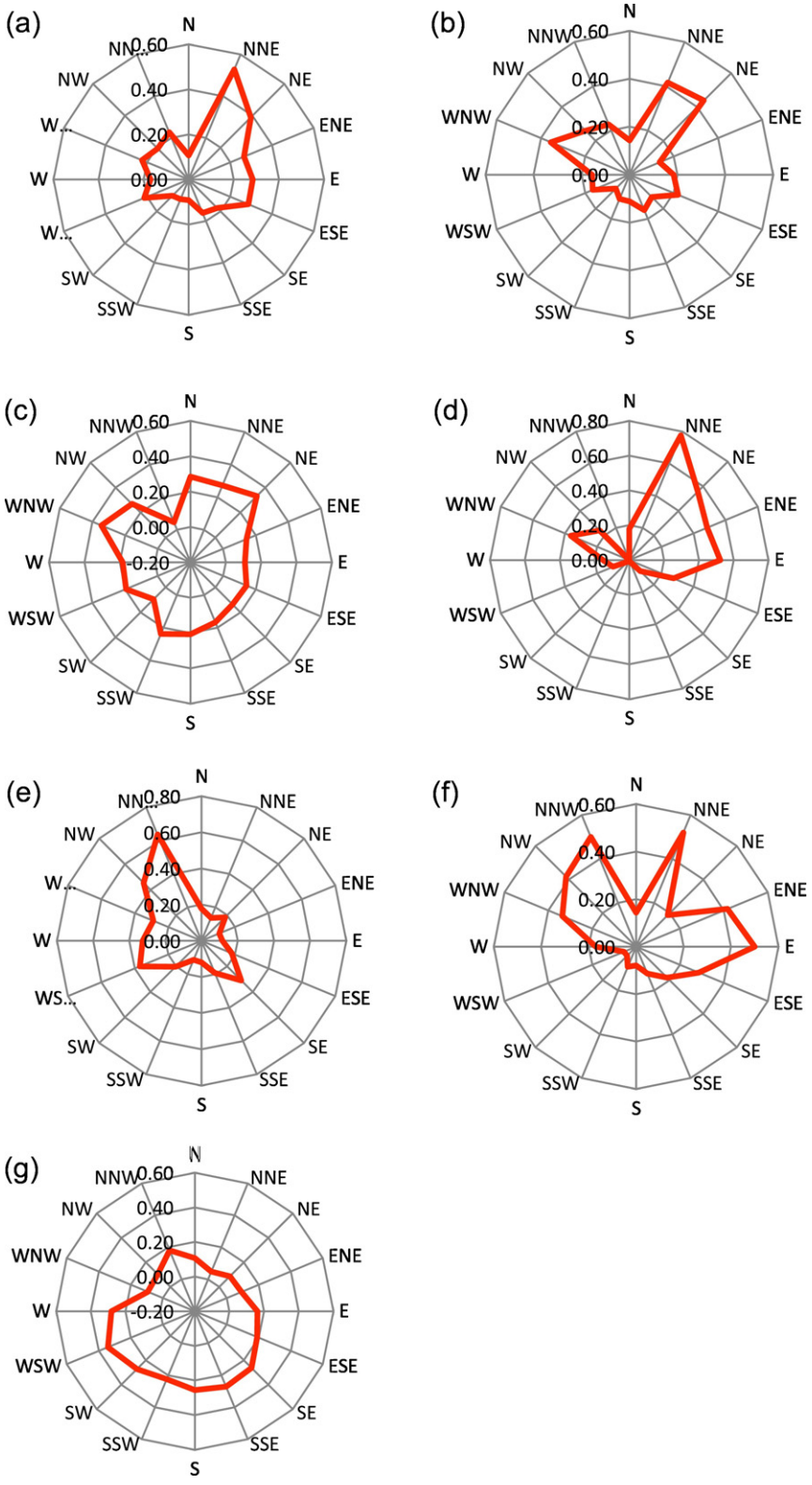

Fig. 4. Likely source areas of (a) secondary inorganic (b) coal combustion (c) traffic-related emissions (d) iron ore and steel industry (e) oil combustion (f) non-ferrous metallurgy and (g) soil dust aerosol in study area using CPF analysis.

of nearby harbors in central Taiwan are referred to this contribution source.

Factor 6 is associated with non-ferrous metallurgy for $PM_{2.5}$ due to the high percentages of Ba (87.7%), Sr (55.4%), Se (45.9%), Cr (29.4%) and Al (29.2%) as shown in Fig. 2. Enriched Sr, Se, Cr and Al have been reported for non-ferrous metallurgy source (Kuo et al., 2007; Querol et al., 2007; Viana et al., 2008). Kuo et al. (2007) indicated that a secondary aluminum smelter was the main source with high concentrations of Al, Cr and Sr in southern Taiwan. The high percentage of Se, Sr, and Ba could also be attributed to glass production (Chen et al., 2015). A high content of Ba was also observed in this source, while it was considered as resuspended dust (Amato et al., 2011). In Fig. 3f, a good correlation

between the time series contribution of non-ferrous metallurgy and the temporal concentration for Sr ($R^2 = 0.60$) was observed. In this factor, the CPF plot shows a high contribution in the direction of NNW, NNE, and E (50% on average) (Fig. 4f) where several non-ferrous smelters and glass manufacturing facility are located (e.g. the Chang-Bin industrial park (NE of study area) and Yunlin County (E of study area)).

Factor 7 is referred to as soil dust, which is typically characterized by high contributions of crustal elements, such as Fe (39.2%), Al (28.0%), Na (30.8%) and Ti (41.3%). Ti, Fe, Na and Al, all of which are major crustal elements, are known as the main components of the airborne soil and road dust (Lough et al., 2005; Zhang et al., 2013). In Fig. 3g, the time series contribution for soil dust presents a good correlation with the temporal concentration of Ti ($R^2 = 0.74$). The CPF plot (Fig. 4g) exhibits wind directions from WSW to SE with a high probability, indicating that local sources of riverbed dust from the Choshui River (the largest one in Taiwan), agricultural field dust, resuspended road dust and nearby ocean are relevant. Asian dust storm triggered by cold air masses passing through northern China and Mongolia also could enhance aerosol concentrations over Taiwan (particularly the north and central regions) during the winter and spring. The characteristic ratio of Fe/Al (=0.6) is often used to recognize its influence (Hsu et al., 2013, and references therein). Given our data of 0.12, 0.13 and 0.26 for the ratio of Fe/Al in the spring, summer and winter, respectively, the influence of the Asian dust storms on the study area during the sampling periods could be ignored.

3.3. Health impact and control strategy

Table 3 shows the average estimates of the relative risk (RR, 95% confidence interval) with a daily elevation of $PM_{2.5}$ of 10 μg m^{-3} in exposure by region and season. The average pooled estimate of the overall RR (yearly overall regions) was 1.020 (95% CI = 1.015–1.025; p-value < 0.001). Yearly, a slight difference in RR estimates was observed among regions. By season, we found that the pooled estimate of the RR was slightly higher in the winter and autumn for all regions, followed by spring and summer. The point estimates of the increased RR on average for the overall regions across seasons were within the range of 1–2% in this study. Our result is similar to that of previous studies. For instance, Dominici et al. (2006) reported that the percentage change in respiratory hospitalization per 10 μg m^{-3} increase in $PM_{2.5}$ for the elderly aged ≥65 years across 204 counties was 0.92% (95% CI = 0.41–1.82). Bell et al. (2008) estimated the increase in the respiratory hospital admission rate per 10 μg m^{-3} increase in $PM_{2.5}$ at log 0 for 202 US counties from 1999 to 2005 to be 0.22% for all seasons and 1.05% for winter nationwide. Zanobetti et al. (2009) indicated an increase of 2.07% (95% CI = 1.2–2.95) in respiratory admissions per 10 μg m^{-3} increase in 2-day averaged $PM_{2.5}$ for US citizens aged ≥65 years for the years 2000–2003. Wong et al. (2006) also reported an increase of 2.1% in respiratory visits in eight districts of Hong Kong from 2000 to 2002. The seasonal heterogeneities in the RR estimates within the Changhua and Yunlin regions might be explained by the $PM_{2.5}$ level, such as physical properties and chemical constituents. Although the mass concentrations and chemical constituents of $PM_{2.5}$ corresponding to the identified sources remaining a significant cause of health impacts have been reported (Bell et al., 2008, 2010; Dominici et al., 2006; Thurston et al., 2016), other factors may also be relevant. The effects of the $PM_{2.5}$ mass and its chemical constituents on the spatial heterogeneity of the RR estimates could be probably ruled out in the study area due to the low spatial variation in those concentrations within the region.

Using Eqs. (3) and (4), we found that the attributable number of cases per 10 μg m^{-3} increment in $PM_{2.5}$ (D_{10}) for respiratory physician visits was 7248 overall yearly. The highest respiratory physician visits (n = 2123) were observed in the winter while the lowest count (n = 1224) was presented in the summer. Based on our current measurements of the $PM_{2.5}$ concentrations (i.e., annual = 25.0 μg m^{-3}; winter = 36.1 μg m^{-3}) shown in Table 1, we can expect the increases of factors of 2.5 and 3.6 in respiratory physician visits for annual (n = 18.097) and winter (n = 7664), respectively, among the elderly aged ≥65 years in Changhua and Yunlin Counties. When the target level of 15 μg m^{-3} in the annual concentration of $PM_{2.5}$ set by the Taiwan EPA for 2020 is achieved from the current level (25 μg m^{-3}), we estimate a potential benefit of a 2% decrease in the total burden of respiratory physician visits for the elderly. In particular, in the winter, which presents relatively higher levels of $PM_{2.5}$ (36.1 μg m^{-3}) and the estimated RR (increase of 2.1%), it is more important to specify the main origins of $PM_{2.5}$ emissions for sufficient controls in this season. Here we found coal combustion, the iron ore and steel industry and non-ferrous metallurgy to be the top three contributors accounting for 55% of the total $PM_{2.5}$ and 70% of the primary $PM_{2.5}$ (excluding secondary inorganic aerosol) in the winter. Not only the mass concentration of $PM_{2.5}$, but also the toxic metals, i.e., As, Cd, Sb, and Pb, derived from those emission sources can be significantly reduced. Thus, those three emission sources are suggested to be first controlled. In addition, we found that fossil-fuel combustion-related activities contributed nearly 94.8% to the $PM_{2.5}$. These activities include source factors of the secondary inorganic aerosols, coal combustion, traffic-related emissions, the iron ore and steel industry, oil combustion and non-ferrous metallurgy. Using high-quality coal and oil with well-managed low sulfur levels is recommended. In general, the PM and its precursor concentrations can be abated in coal-combustion-generated emissions by adjusting the air-fuel ratio, de-rating engines to limit the power output, and using end-of-pipe control devices. In addition, advanced abatement technologies should be employed for $PM_{2.5}$ and precursor gas, and the coal-fired power plants should be upgraded to natural gas-fired power plants. Although our measured data did not directly link to the adverse health outcome of populations to toxic compounds, as found in previous studies (Bell et al., 2014; Thurston et al., 2016), because of insufficient exposure and health information, our study may aid efforts on health protection from the aspect of particles mass reduction focusing on the specific season and source. A systematic review of $PM_{2.5}$-sized compounds and health indicated that SO_4^{2-}, NH_4^+, OC and EC were positively associated with increased all-cause, cardiovascular and respiratory mortality

Table 3
Estimates of the relative risk (RR and 95% confidence interval (CI)) for respiratory physician visits per 10 μg m^{-3} increment in $PM_{2.5}$ by township and season.

Regions	Yearly[a]			Spring			Summer			Autumn			Winter		
	RRs	95% CI		RRs	95% CI		RRs	95% CI		RRs	95% CI		RRs	95% CI	
All regions[b]	1.020	1.015	1.025	1.019	1.01	1.027	1.017	0.998	1.036	1.023	1.009	1.036	1.021	1.011	1.031
Erlin	1.019	1.002	1.037	1.014	0.986	1.044	0.987	0.930	1.046	1.039	1.001	1.079	1.020	0.991	1.049
Changhua	1.030	1.011	1.049	1.007	0.973	1.041	1.046	0.986	1.110	1.052	1.015	1.090	1.029	1.003	1.055
Hsienhsi	1.019	1.012	1.026	1.022	1.033	1.012	1.032	1.008	1.057	1.009	0.996	1.022	1.019	1.010	1.029
Douliou	1.024	1.016	1.032	1.020	1.007	1.034	1.005	0.977	1.034	1.023	1.006	1.041	1.031	1.019	1.044
Lunbei	0.983	0.958	1.008	0.969	0.923	1.016	0.956	0.875	1.045	1.010	0.959	1.065	0.982	0.943	1.022
Taixi	1.016	0.993	1.040	1.019	0.972	1.069	1.025	0.939	1.118	1.038	0.986	1.092	1.005	0.972	1.039

[a] Pooled estimates of RRs from four seasons.
[b] Pooled estimates of RRs from six townships.

(Atkinson et al., 2015). Those addressed compounds account for >50% of the total $PM_{2.5}$ mass in our study, so our proposed control measures might be effective to protect health in the study area.

The available health data of the NHIRD only provides morbidity information for the Taiwanese population; it does not allow us to analyze the mortality associated with $PM_{2.5}$ exposure, in spite of the strong evidence of the relationship between $PM_{2.5}$ exposure and mortality risk (Dockery et al., 1993; Dominici et al., 2006; Pope et al., 2002). The use of health data for the years of 2006–2011 in this study is limited to personal information on smoking, lifestyle, and indoor/outdoor activities, which does not allow us to account for such confounding factors. We also assumed that the contributors of $PM_{2.5}$ and resultant seasonal patterns in 2006–2011 are similar to those in more recent years (2014–2015), although the structures of the industry, traffic, and population and meteorological conditions at/near the study area may somewhat differ between two periods.

4. Conclusion

Our study provided comprehensive results on the chemical constituents of $PM_{2.5}$ with spatial and seasonal variations. While a low spatial contract in $PM_{2.5}$ and associated chemical concentrations was observed in a residential area near industrial complexes in central Taiwan, the seasonal variation showed that those were higher in the winter than in the spring or summer. Our result also clarified the contributions of potential $PM_{2.5}$ sources in this area. In addition to nearby oil refinery plants associated with oil combustion, coal combustion, traffic-related emissions, the iron ore and steel industry and non-ferrous metallurgy are other potentially important contributors to the ambient $PM_{2.5}$. An increment in the daily exposure to $PM_{2.5}$ of 10 μg m^{-3} could result in a 1–2% increase in respiratory physician visits for elderly aged ≥65 years, in particular in the winter. We prioritized coal combustion, the iron ore and steel industry and non-ferrous metallurgy as the top three contributors of $PM_{2.5}$ during the winter for controls, considering the effective reduction of the health burdens, i.e., respiratory physician visits.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.02.212.

References

Alleman, L.Y., Lamaison, L., Perdrix, E., Robache, A., Galloo, J.C., 2010. PM_{10} metal concentrations and source identification using positive matrix factorization and wind sectoring in a French industrial zone. Atmos. Environ. 96, 612–625.

Amato, F., Viana, M., Richard, A., Furger, M., Prévôt, A.S.H., Nava, S., Lucarelli, F., Bukowiecki, N., Alastuey, A., Reche, C., Moreno, T., Pandolfi, M., Pey, J., Querol, X., 2011. Size and time-resolved roadside enrichment of atmospheric particulate pollutants. Atmos. Chem. Phys. 11, 2917–2931.

Atkinson, R.W., Mills, I.C., Walton, H.A., Anderson, H.R., 2015. Fine particle components and health—a systematic review and meta-analysis of epidemiological time series studies of daily mortality and hospital admissions. J. Expo. Anal. Environ. Epidemiol. 25, 208–214.

Belis, C.A., Karagulian, F., Larsen, B.R., Hopke, P.K., 2013. Critical review and meta-analysis of ambient particulate matter source apportionment using receptor models in Europe. Atmos. Environ. 69, 94–108.

Bell, M.L., Ebisu, K., Peng, R.D., Walker, J., Samet, J.M., Zeger, S.L., Dominici, F., 2008. Seasonal and regional short-term effects of fine particles on hospital, admissions in 202 US counties, 1999–2005. Am. J. Epidemiol. 168, 1301–1310.

Bell, M.L., Belanger, K., Ebisu, K., Gent, J.F., Lee, H.J., Koutrakis, P., Leaderer, B.P., 2010. Prenatal exposure to fine particulate matter and birth weight: variations by particulate constituents and sources. Epidemiology 21, 884–891.

Bell, M.L., Ebisu, K., Leaderer, B.P., Gent, J.F., Lee, H.J., Koutrakis, P., Wang, Y., Dominici, F., Peng, R.D., 2014. Associations of $PM_{2.5}$ constituents and sources with hospital admissions: analysis of four counties in Connecticut and Massachusetts (USA) for persons ≥65 years of age. Environ. Health Perspect. 122, 138–144.

Bencs, L., Ravindra, K., de Hoog, J., Rasoazanany, E.O., Deutsch, F., Bleux, N., Berghmans, P., Roekens, E., Krata, A., Van Grieken, R., 2008. Mass and ionic composition of atmospheric fine particles over Belgium and their relation with gaseous air pollutant. J. Environ. Monit. 10, 1148–1157.

Boldo, E., Medina, S., LeTertre, A., Hurley, F., Mucke, H.G., Ballester, F., Aguilera, I., Eilstein, D., 2006. Apheis: health impact assessment of long-term exposure to $PM_{2.5}$ in 23 European cities. Eur. J. Epidemiol. 21, 449–458.

Cascio, W.E., Katwa, L.C., Linn, W.S., Stram, D.O., Zhu, Y., Cascio, J.L., Hinds, W.C., 2009. Effects of vehicle exhaust in aged adults riding on Los Angeles freeways. Am. J. Respir. Crit. Care Med. 179, A1175.

Chen, L.C., Lippmann, M., 2009. Effects of metals within ambient air particulate matter (PM) on human health. Inhal. Toxicol. 21, 1–31.

Chen, Y.C., Hsu, C.Y., Lin, S.L., Chang-Chien, G.P., Chen, M.J., Fang, G.C., Chiang, H.C., 2015. Characteristics of concentrations and metal compositions for $PM_{2.5}$ and $PM_{2.5-10}$ in Yunlin County, Taiwan during air quality deterioration. Aerosol Air Qual. Res. 15, 2571–2583.

Chen, Y.C., Chiang, H.C., Hsu, C.Y., Yang, T.T., Chen, N.T., Lin, T.Y., Chen, M.J., Wu, Y.S., 2016. Ambient PM2.5-bound polycyclic aromatic hydrocarbons (PAHs) in Changhua of central Taiwan: seasonal variation, source apportionment and cancer risk assessment. Environ. Pollut. 218, 372–382.

Cheng, W.L., Pai, J.L., Tsuang, B.J., Chen, C.L., 2001. Aerosol and air quality research, 9: 421–434, 2009 433 synoptic patterns in relation to ozone concentrations in west-central Taiwan. Meteorog. Atmos. Phys. 78, 11–21.

Cheng, K., Wang, Y., Tian, H., Gao, X., Zhang, Y., Wu, X., Zhu, C., Gao, J., 2015. Atmospheric emission characteristics and control policies of five precedent-controlled toxic heavy metals from anthropogenic sources in China. Environ. Sci. Technol. 49, 1206–1214.

Chou, C.C.K., Lee, C.T., Cheng, M.T., Yuan, C.S., Chen, S.J., Wu, Y.L., Hsu, W.C., Lung, S.C., Hsu, S.C., Lin, C.Y., Liu, S.C., 2010. Seasonal variation and spatial distribution of carbonaceous aerosols in Taiwan. Atmos. Chem. Phys. 10, 9563–9578.

Comrie, A.C., 1994. A synoptic climatology of rural ozone pollution at three forest sites in Pennsylvania. Atmos. Environ. Health Perspect. 28, 1601–1614.

Contini, D., Cesari, D., Genga, A., Siciliano, M., Ielpo, P., Guascito, M., Conte, M., 2014. Source apportionment of size-segregated atmospheric particles based on the major water-soluble components in Lecce (Italy). Environ. Sci. Technol. 472, 248–261.

Dall'Osto, M., Querol, X., Amato, F., Karanasiou, A., Lucarelli, F., Nava, S., Calzolai, G., Chiari, M., 2013. Hourly elemental concentrations in $PM_{2.5}$ aerosols sampled simultaneously at urban background and road site during sapuss-diurnal variations and PMF receptor modelling. Atmos. Chem. Phys. 13, 4375–4392.

Dockery, D.W., Pope, C.A., Xu, X., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris, B.G., Speizer, F.E., 1993. An association between air pollution and mortality in six U.S. cities. N. Engl. J. Med. 329, 1753–1759.

Dominici, F., Peng, R.D., Bell, M.L., Pham, L., McDermott, A., Zeger, S.L., Samet, J.M., 2006. Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases. JAMA 295, 1127–1134.

Franklin, M., Koutrakis, P., Schwartz, J., 2008. The role of particle composition on the association between $PM_{2.5}$ and mortality. Epidemiology 19, 680–689.

Gehring, U., Beelen, R., Eeftens, M., Hoek, G., de Hoogh, K., de Jongste, J.C., Keuken, M., Koppelman, G.H., Meliefste, K., Oldenwening, M., Postma, D.S., van Rossem, L., Wang, M., Smit, H.A., Brunekreef, B., 2015. Particulate matter composition and respiratory health: the Piama birth cohort study. Epidemiology 26, 300–309.

Gu, J., Bai, Z., Liu, A., Wu, L., Xie, Y., Li, W., Dong, H., Zhang, X., 2010. Characterization of atmospheric organic carbon and element carbon of $PM_{2.5}$ and PM_{10} at Tianjin,China. Aerosol. Air Qual. Res. 10, 167–176.

Han, J.S., Moon, K.J., Lee, S.J., Kim, Y.J., Ryu, S.Y., Cliff, S.S., Yi, S.M., 2006. Size-resolved source apportionment of ambient particles by positive matrix factorization at gosan background site in east Asia. Atmos. Chem. Phys. 6, 211–223.

Heo, J.B., Hopke, P.K., Yi, S.M., 2009. Source apportionment of $PM_{2.5}$ in Seoul, Korea. Atmos. Chem. Phys. 9, 4957–4971.

Hsu, S.C., Tsai, F., Lin, F.J., Chen, W.N., Shiah, F.K., Huang Jr., C., Chan, C.Y., Chen, C.C., Liu, T.H., Chen, H.Y., 2013. A super Asian dust storm over the East and South China Seas: disproportionate dust deposition. J. Geophys. Res. Atmos. 118, 7169–7181.

Hsu, C.Y., Chiang, H.C., Lin, S.L., Chen, M.J., Lin, T.Y., Chen, Y.C., 2016. Elemental characterization and source apportionment of PM_{10} and $PM_{2.5}$ in the western coastal area of central Taiwan. Sci. Total Environ. 541, 1139–1150.

Huang, F., Li, X., Wang, C., Xu, Q., Wang, W., Luo, Y., Tao, L., Gao, Q., Chen, S., Cao, K., Liu, L., Gao, N., 2015. PM2.5 spatiotemporal variations and the relationship with meteorological factors during 2013–2014 in Beijing, China. PLos One 10, e0141642.

Kang, C.M., Kang, B.W., Lee, H.S., 2006. Source identification and trends in concentrations of gaseous and fine particulate principal species in Seoul, South Korea. J. Air Waste Manage. Assoc. 56, 911–921.

Kim, E., Hopke, P.K., Edgerton, E.S., 2003. Source identification of Atlanta aerosol by positive matrix factorization. J. Air Waste Manage. Assoc. 53, 731–739.

Kim, J.Y., Song, C.H., Ghim, Y.S., Won, J.G., Yoon, S.C., Carmichael, G.R., Woo, J.H., 2006. An investigation on NH_3 emissions and particulate NH^+_4–NO^-_3 formation in East Asia. Atmos. Environ. 40, 2139–2150.

Kumar, U., Prakash, A., Jain, V.K., 2008. A photochemical modeling approach to investigate O_3 sensitivity to NOx and VOCs in the urban atmosphere of Delhi. Aerosol Air Qual. Res. 8, 147–159.
Kunzli, N., Kaiser, R., Medina, S., Studnicka, M., Chanel, O., Filliger, P., Herry, M., Horak Jr., F., Puybonnieux-Texier, V., Quenel, P., Schneider, J., Seethaler, R., Vergnaud, J.C., Sommer, H., 2000. Public-health impact of outdoor and traffic-related air pollution: a European assessment. Lancet 356, 795–801.
Kuo, S.C., Hsiehb, L.Y., Tsaic, C.H., Tsai, Y.I., 2007. Characterization of $PM_{2.5}$ fugitive metal in the workplaces and the surrounding environment of a secondary aluminum smelter. Atmos. Environ. 41, 6884–6900.
Kuo, C.Y., Lin, C.Y., Huang, L.M., Wang, S., Shieh, P.F., Lin, Y.R., Wang, J.Y., 2010. Spatial variations of the aerosols in river–dust episodes in central Taiwan. J. Hazard. Mater. 179, 1022–1030.
Kuo, C.Y., Lin, Y.R., Chang, S.Y., Lin, C.Y., Chou, C.H., 2013. Aerosol characteristics of different types of episode. Environ. Monit. Assess. 185, 9777–9787.
Lee, J.H., Hopke, P.K., 2006. Apportioning Sources of $PM_{2.5}$ in St. Luis, Mo using speciation trends network data. Atmos. Environ. 40, S360–S377.
Liang, C., Duan, F., He, K., Ma, Y., 2016. Review on recent progress in observations, source identifications and countermeasures of $PM_{2.5}$. Environ. Int. 86, 150–170.
Liao, H.T., Chou, C.C.K., Chow, J.C., Watson, J.G., Hopke, P.K., Wu, C.F., 2015. Source and risk apportionment of selected VOCs and $PM_{2.5}$ species using partially constrained receptor models with multiple time resolution data. Environ. Pollut. 205, 121–130.
Lin, C.Y., Liu, S.C., Chou, C.C.K., Liu, T.H., Lee, C.T., 2004. Long-range transport of Asian dust and air pollutants to Taiwan. Terr. Atmos. Ocean. Sci. 15, 759–784.
Lin, Y.C., Tsai, C.J., Wu, Y.C., Zhang, R., Chi, K.H., Huang, Y.T., Lin, S.H., Hsu, S.C., 2015. Characteristics of trace metals in traffic-derived particles in Hsuehshan Tunnel, Taiwan: size distribution, potential source, and fingerprinting metal ratio. Atmos. Chem. Phys. 15, 4117–4130.
Lough, G.C., Schauer, J.J., Park, J.S., Shafer, M.M., Deminter, J.T., Weinstein, J.P., 2005. Emissions of metals associated with motor vehicle roadways. Environ. Sci. Technol. 39, 826–836.
Manoli, E., Voutsa, D., Samara, C., 2002. Characterization and source identification/apportionment of fine and coarse air particles in Thessaloniki, Greece. Atmos. Environ. 36, 949–961.
Mokhtara, M.M., Taiba, R.M., Hassima, M.H., 2014. Understanding selected trace elements behavior in a coal–fired power plant in Malaysia for assessment of abatement technologies. J. Air Waste Manage. Assoc. 64, 867–878.
Mooibroek, D., Schaap, M., Weijers, E., Hoogerbrugge, R., 2011. Source apportionment and spatial variability of $PM_{2.5}$ using measurements at five sites in the netherlands. Atmos. Environ. 45, 4180–4191.
Pacyna, E.G., Pacyna, J.M., Fudala, J., Strzelecka-Jastrzab, E., Hlawiczka, S., Panasiuk, D., Nittere, S., Preggere, T., Pfeiffere, H., Friedrich, R., 2007. Current and future emissions of selected heavy metals to the atmosphere from anthropogenic sources in Europe. Atmos. Environ. 41, 8557–8566.
Pope, C.I., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K., Thurston, G., 2002. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. JAMA 287, 1132–1141.
Querol, X., Viana, M., Alastuey, A., Amato, F., Moreno, T., Castillo, S., Pey, J., de la Rosa, J., Sánchez de la Campa, A., Artíñano, B., Salvador, P., García Dos Santos, S., Fernández-Patier, R., Moreno-Grau, S., Negral, L., Minguillón, M.C., Monfort, E., Gil, J.I., Inza, A., Ortega, L.A., Santamaría, J.M., Zabalza, J., 2007. Source origin of trace elements in PM from regional background, urban and industrial sites of Spain. Atmos. Environ. 41, 7219–7231.
Seinfeld, J.H., Pandis, S.N., 1998. Atmospheric Chemistry and Physics. Willey/Interscience.
Shafer, M.M., Toner, B.M., Overdier, J.T., Schauer, J.J., Fakra, S.C., Hu, S., Herner, J.D., Ayala, A., 2012. Chemical speciation of vanadium in particulate matter emitted from diesel vehicles and urban atmospheric aerosols. Environ. Sci. Technol. 46, 189–195.
Song, Y., Zhang, Y.H., Xie, S.D., Zeng, L.M., Zheng, M., Salmon, L.G., Shao, M., Slanina, S., 2006. Source apportionment of $PM_{2.5}$ in Beijing by positive matrix factorization. Atmos. Environ. 40, 1526–1537.
Stortini, A.M., Freda, A., Cesari, D., Cairns, W.R.L., Contini, D., Barbante, C., Prodi, F., Cescon, P., Gambaro, A., 2009. An evaluation of the $PM_{2.5}$ trace elemental composition in the Venice Lagoon area and an analysis of the possible sources. Atmos. Environ. 43, 6296–6304.
Tai, A.P.K., Mickley, L.J., Jacob, D.J., 2010. Correlations between fine particulate matter ($PM_{2.5}$) and meteorological variables in the United States: Implications for the sensitivity of $PM_{2.5}$ to climate change. Atmos. Environ. 44, 3976–3984.
Tao, J., Gao, J., Zhang, L., Zhang, R., Che, H., Zhang, Z., Zhang, R., Che, H., Zhang, Z., Lin, Z., Jing, J., Cao, J., Hsu, S.C., 2014. $PM_{2.5}$ pollution in a mega city of southwest China: source apportionment and implication. Atmos. Chem. Phys. 14, 8679–8699.
Thurston, G.D., Burnett, R.T., Turner, M.C., Shi, Y., Krewski, D., Lall, R., Ito, K., Jerrett, M., Gapstur, S.M., Diver, W.R., Pope, C.A., 2016. Ischemic heart disease mortality and long-term exposure to source-related components of U.S. fine particle air pollution. Environ. Health Perspect. 124, 785–794.
Tian, H.Z., Wang, Y., Xue, Z.G., Cheng, K., Qu, Y.P., Chai, F.H., Hao, J.M., 2010. Trend and characteristics of atmospheric emissions of Hg, As, and Se from coal combustion in China, 1980–2007. Atmos. Chem. Phys. 10, 11905–11919.
Tian, H., Zhao, D., Cheng, K., Lu, L., He, M., Hao, J., 2012. Anthropogenic atmospheric emissions of antimony and its spatial distribution characteristics in China. Environ. Sci. Technol. 46, 3973–3980.
Tsai, Y.I., Chen, C.L., 2006. Atmospheric aerosol composition and source apportionments to aerosol in southern Taiwan. Atmos. Environ. 40, 4751–4763.
Tsai, J.H., Lin, K.H., Chen, C.Y., Ding, J.Y., Choa, C.G., Chiang, H.L., 2007. Chemical constituents in particulate emissions from an integrated iron and steel facility. J. Hazard. Mater. 147, 111–119.
Viana, M., Kuhlbusch, T.A.J., Querol, X., Alastuey, A., Harrison, R.M., Hopke, P.K., Winiwarter, W., Vallius, A., Szidat, S., Prevot, A.S.H., Hueglin, C., Bloemen, H., Wahlin, P., Vecchi, R., Miranda, A.I., Kasper-Giebl, A., Maenhaut, W., Hitzenberger, R., 2008. Source apportionment of particulate matter in Europe: a review of methods and results. J. Aerosol Sci. 39, 827–849.
Vukovich, F.M., 1994. Boundary layer ozone variations in the eastern United States and their associations with meteorological variations: long-term variations. J. Geophys. Res. 99, 16839–16850.
Wang, W.C., Chen, K.S., 2008. Modeling and analysis of source contribution of PM_{10} during severe pollution events in southern Taiwan. Aerosol Air Qual. Res. 8, 319–338.
Wang, J., Ogawa, S., 2015. Effects of meteorological conditions on $PM_{2.5}$ concentrations in Nagasaki, Japan. Int. J. Environ. Res. Public Health 12, 9089–9101.
Wang, C.F., Chin, C.J., Chiang, P., 1998. Multi-element analysis of suspended particulates collected with a betagauge monitoring system by ICP atomic emission spectrometry and mass spectrometry. Anal. Sci. 14, 763–768.
Wang, Y.F., Huang, K.L., Li, C.T., Mi, H.H., Luo, J.H., Tsai, P.J., 2003. Emissions of fuel metals content from a diesel vehicle engine. Atmos. Environ. 37, 4637–4643.
WHO European Centre for Health Policy, 2000. Health impact assessment: main concepts and suggested approach. The Gothenburg Consensus Paper. WHO Regional Office for Europe, Copenhagen.
Wong, T.W., Tam, W., Yu, I.T.S., Wun, Y.T., Wong, A.H.S., Wong, C.M., 2006. Association between air pollution and general practitioner visits for respiratory diseases in Hong Kong. Thorax 61, 585–591.
Yang, H.H., Chen, H.W., Chi, T.W., Chuang, P.Y., 2008. Analysis of atmospheric ozone concentration trends as measured by eighth highest values. Aerosol Air Qual. Res. 8, 308–318.
Yongming, H., Peixuan, D., Junji, C., Posmentier, E.S., 2006. Multivariate analysis of heavy metal contamination in urban dusts of Xi'an central China. Sci. Total Environ. 335, 176–186.
Zanobetti, A., Franklin, M., Koutrakis, P., Schwartz, J., 2009. Fine particulate air pollution and its components in association with cause-specific emergency admissions. Environ. Health 8, 58.
Zhang, Y.P., Wang, X.F., Chen, H., Yang, X., Chen, J.M., Allen, J.O., 2009. Source apportionment of lead-containing aerosol particles in Shanghai using single particle mass spectrometry. Chemosphere 74, 501–507.
Zhang, R.J., Jing, J., Tao, J., Hsu, S.C., Wang, G., Cao, J., 2013. Chemical characterization and source apportionment of $PM_{2.5}$ in Beijing: seasonal perspective. Atmos. Chem. Phys. 13, 7053–7074.