



Source Characterization and Apportionment of PM₁₀, PM_{2.5} and PM_{0.1} by Using Positive Matrix Factorization

Balakrishnaiah Gugamsetty¹, Han Wei¹, Chun-Nan Liu¹, Amit Awasthi¹, Shih-Chieh Hsu², Chuen-Jinn Tsai^{1*}, Gwo-Dong Roam², Yue-Chuen Wu², Chung-Fang Chen²

¹ *Institute of Environmental Engineering, National Chiao Tung University, Hsinchu 300, Taiwan*

² *Research Center for Environmental Changes, Academia Sinica, Taipei 115, Taiwan*

³ *Environmental Analysis Laboratory, Environmental Protection Administration, Zhongli 320, Taiwan*

ABSTRACT

Ambient Particulate Matters (PM₁₀, PM_{2.5} and PM_{0.1}) were investigated at Shinjung station in New Taipei City, Taiwan. Samples were collected simultaneously using a dichotomous sampler (Andersen Model SA-241) and a MOUDI (MSP Model 110) over a 24-h period from May 2011 to November 2011 at Shinjung station. Samples were analyzed for metallic trace elements using ion coupled plasma mass spectroscopy (ICP-MS) and ionic compounds by ion chromatography (IC). The average concentrations of PM₁₀, PM_{2.5} and PM_{0.1} were found to be 39.45 ± 11.58 , 21.82 ± 7.50 and $1.42 \pm 0.56 \mu\text{g}/\text{m}^3$, respectively. Based on the chemical information, positive matrix factorization (PMF) was used to identify PM sources. A total of five source types were identified, including soil dust, vehicle emissions, sea salt, industrial emissions and secondary aerosols, and their contributions were estimated using PMF. The crustal enrichment factors (EF) were calculated using Al as a reference for the trace metal species to identify the sources. Conditional probability functions (CPF) were computed using wind profiles and factor contributions. The results of CPF analysis were used to identify local point sources. The results suggest a competitive relationship between anthropogenic and natural source processes over the monitoring station.

Keywords: Positive matrix factorization; Enrichment factor analysis; Conditional probability function analysis; PM₁₀; PM_{2.5}; PM_{0.1}.

INTRODUCTION

The role of particulate matter (PM) in climate change has long been recognized (IPCC, 2007), and aerosols can also adversely affect human health via inhalation, especially in the urban environment (Khan *et al.*, 2010; Kim *et al.*, 2011). Epidemiological studies show different associations between adverse health effects and particles with aerodynamic diameters of less than 2.5 μm (PM_{2.5}) and less than 10 μm (PM₁₀) (Barnpadimos *et al.*, 2011; McBride *et al.*, 2011). The sources of PM_{2.5} and PM₁₀ are different, and include a wide range of natural phenomena and human activities. PM₁₀ particles mainly originate from sea salt, soil dust re-suspension, construction/demolition, non-exhaust vehicle emissions, and industrial fugitives, whereas PM_{2.5} and PM_{0.1} particles are mainly produced by combustion processes, forest fires and transformation of gaseous species. The lifetime of smaller size particles (PM_{2.5} and PM_{0.1}) can range

from days to weeks, while bigger particles (PM₁₀) have a lifetime of hours to days. Atkinson *et al.* (2010) investigated the associations between PM fractions and respiratory/cardiovascular mortality and morbidity in London, and the results were largely dependent on the day lag examined and the type of health outcome. The most significant effects were recorded for 1-day lagged respiratory hospital admissions, which was comparable to the corresponding effect of PM_{2.5}.

Several studies show that ambient particulate pollution is associated with certain health and environmental effects (Choosong *et al.*, 2010; Ning *et al.*, 2010; Wang *et al.*, 2012). Trace elements are important components of aerosols, and industrial, residential, and traffic related activities have resulted in a substantial increase in trace metals (e.g., Cu, Pb, Zn, Cd, Ni etc.) in the atmosphere. The organic components of atmospheric aerosols play an important role in particles with an aerodynamic diameter of less than 1 μm (fine mode). Aside from high concentrations of coarse mineral particles, PM_{2.5} can be observed when Asian dust (AD) approaches Taiwan (Chang *et al.*, 2010). Emissions from mega-cities are also a source of pollutants to other parts of the world through their long range transport. A reliable quantitative estimation of PM_{2.5} and PM₁₀ is thus

* Corresponding author. Tel.: +886-3-5731880;
Fax: +886-3-5727835
E-mail address: cjtsai@mail.nctu.edu.tw

needed to support the implementation of air quality modeling studies (Sahu *et al.*, 2011).

The identification of emission sources and quantification of their contributions to the ambient concentration of pollutants has been a major focus of urban air quality research. The development and application of improved tools is thus required for the identification and apportionment of atmospheric aerosols. Receptor modeling offers a way to achieve this by measuring the concentration of pollutants at a sampling site (Hopke, 1991). One type of receptor model is the multivariate model, which was recently improved significantly due to a new approach called positive matrix factorization (PMF) developed by Paatero and colleagues (Paatero and Tapper, 1993, 1994; Paatero, 1997) using a least squares approach. PMF solves the problems arising in factor analysis by integrating non-negativity constraints into the optimization process, and utilizing the error estimates for each data value as point-by-point weights (Begum *et al.*, 2004). PMF provides a flexible modeling approach and allows effective use of the information in the data, and has been applied successfully to receptor modeling around the world (Polissar *et al.*, 1998; Kim *et al.*, 2004; Lee and

Hopke, 2006; Karanasiou *et al.*, 2009).

The present study thus utilizes PMF to identify the source profiles and apportionment of PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ particles, and to estimate the mass contributions of source factors from different wind directions using the Conditional Probability Function (CPF) at Shinjung station, Taiwan.

METHODOLOGY

Sampling Site Description

Samples were collected at Fu Jen Catholic University campus, which is a four-floor building in Shinjung ($25^{\circ}02'N$, $121^{\circ}25'E$), located in New Taipei City, with the surrounding area shown in Fig. 1. Taipei City has a monsoon-influenced humid subtropical climate, which is slightly different from a true tropical climate. Summers are hot, humid, and accompanied by occasional heavy rainstorms and typhoons, while winters are short, mild and generally very foggy, due to a northeasterly wind. Because of Taiwan's location in the Pacific Ocean, it is affected by the Pacific typhoon season, which occurs between June to October. The sampling station is surrounded by Wugu Industrial Park (3.5 km) and

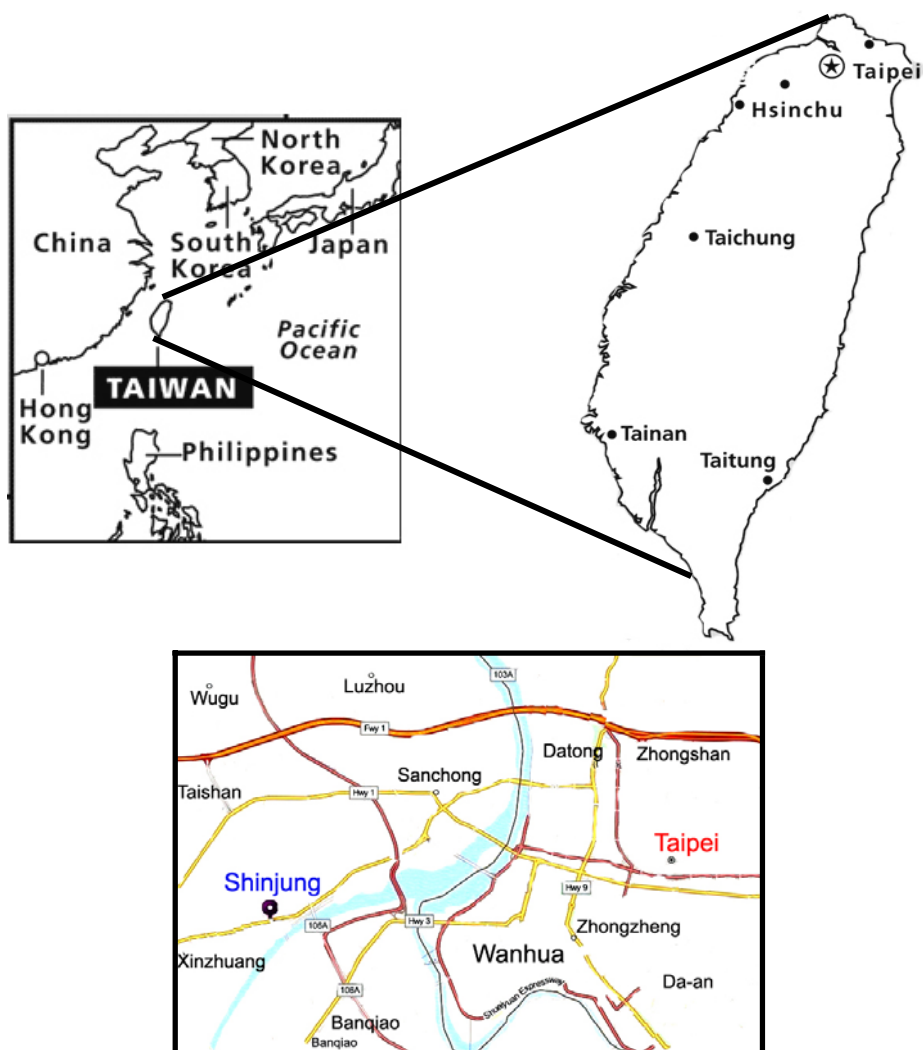


Fig. 1. Location of sampling station in Taiwan.

Taishan Industrial Zone (3.5 km) to the north, Sanchong to the east, Banqiao and Shulin to the south, and Taoyuan County to the west. The traffic flow is heavy during the morning and evening rush hours when commuters as well as neighborhood residents may be exposed to high concentrations of PM. A total of 18 samples were collected during 18 sampling days over a 24-h period from May 2011 to November 2011 at Shinjung station.

Sampling Method and Analysis

PM₁₀, PM_{2.5} and PM_{0.1} samples were collected by using a MOUDI (Model 110, MSP Corp., MN, USA) and a dichotomous samplers (Model SA-241, Andersen Inc., Georgia, USA) operated at flow rates of 30 and 16.7 L/min (Chen *et al.*, 2010), respectively. The MOUDI sampler has 10 size stages, with the nominal cutoff diameters of 18 (inlet), 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18, 0.1 and 0.056 mm (Marple *et al.*, 1991). The substrates used in the study were Teflon (Zefluor P5PJ047, Pall Corp., New York, USA) filters. Stages 0–9 of the MOUDI used silicone grease-coated foils to reduce solid particle bouncing, so that accurate PM_{0.1} samples were obtained by the Teflon filters. The PM samples collected by the silicon grease-coated foils in stages 1–9 of MOUDI (18–0.1 mm) were analyzed gravimetrically but not chemically, because of interference from the grease-coating. The Teflon filters were conditioned for at least 24-h in a temperature and relative humidity controlled room (22 ± 1°C, 40 ± 5% RH) before sampling (Chen *et al.*, 2010). The average mass concentrations of PM₁₀ and PM_{2.5} determined by the MOUDI were in agreement with those obtained from the Dichot, with a relative deviation of < 20% (Chen *et al.*, 2010).

Before chemical analysis, all the Teflon samples were first weighed to determine the mass concentrations of the PM samples. The electrostatic charge of the filters was eliminated by an ionizing air blower (Model CSD-0911, MEISEI, Japan) before weighing. A microbalance (Model CP2P-F, Sartorius, Germany) was used to weigh the filters after being conditioned for at least 24-h in the temperature and relative humidity controlled room. After gravimetric analysis, each Teflon filter was cut equally in half using Teflon coated scissors. One half was analyzed by an ICP-MS (Elan 6100, Perkin Elmer, USA) for the identification of trace elements, while the other was analyzed by an ion chromatograph (IC, Model DX-120, Dionex Corp, Sunnyvale, CA) for ionic species. A Semi-Continuous OC-EC Field Analyzer (Model 4, Sunset Laboratory, Tigard, OR) was used to determine the organic carbon (OC) and elemental carbon (EC) mass concentrations in PM_{2.5}. The OC and EC data were only available for PM_{2.5}, but not for PM₁₀ and PM_{0.1}. The analyzed elements included the major (crustal) elements (Na, Mg, Al, K, Ca, Fe, and Si), sub-major (anthropogenic) elements (Zn, Ni, Cu, Mn, Sr, Ag, Ba, Pb, V, Cr, and Ti, and the ionic elements (F⁻, Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺).

The crustal enrichment factors (EFs) of the trace elements relative to the earth's crust were calculated to indicate the contribution of other sources to the ambient elemental levels (Taylor and McLennan, 1985). The EF of the elements

in PM₁₀, PM_{2.5} and PM_{0.1} mass concentration samples were calculated as:

$$EF = \frac{El_{\text{sample}}/X_{\text{sample}}}{El_{\text{crust}}/X_{\text{crust}}} \quad (1)$$

where El_{sample} and X_{sample} are the element (El) and the reference element (X) mass concentrations in the sample, respectively, and El_{crust} and X_{crust} are the element (El) and the reference element (X) mass concentrations in the upper continental crust, respectively. There is no widely accepted rule for the choice of a reference element, except that it must be a terrigenous one. Al and Fe are the most commonly used elements for this purpose, and in this work the former was used, because it is relatively stable and is not affected by most anthropogenic contaminants.

Source Apportionment by Positive Matrix Factorization (PMF)

Positive Matrix Factorization (PMF v3.0) (USEPA, 2008) was used to identify the contributions of various emission sources. The PMF model is a multivariate factor analysis tool that decomposes the matrix of a speciated sample into two matrices: factor contributions and factor profiles. The PMF v3.0 model requires two input files: one for the measured concentration of the species, and one for the estimated uncertainty of the concentration. PMF is described in greater detail in Paatero and Tapper (1993) and Paatero (1997).

A speciated data set can be viewed as a data matrix X of i by j dimensions, in which i number of samples and j chemical species are measured. The goal of multivariate receptor modeling with PMF is to identify a number of factors (p), the species profile (f) of each source, and the amount of mass (g) contributed by each factor to each individual sample, which is defined as:

$$X_{ij} = \sum_{k=1}^p f_{kj}g_{ik} + e_{ij} \quad (2)$$

where e_{ij} is the residual for each sample/species.

The results are constrained so that no sample can have a negative source contribution. PMF allows each data point to be individually weighed. The objective of the PMF solution is to minimize the object function Q based on the uncertainties (u) as follows (USEPA, 2008):

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left[\frac{x_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right]^2 \quad (3)$$

where x_{ij} are the measured concentrations (in $\mu\text{g}/\text{m}^3$), u_{ij} are the estimated uncertainty values (in $\mu\text{g}/\text{m}^3$), n is the number of samples, m is the number of species and p is the number of sources included in the analysis.

EPA PMF v3.0 accepts two types of uncertainty file: sample-specific and equation-based. The sample-specific uncertainty file provides an estimate of the uncertainty for each sample of each species. While it should have the same dimensions as the concentration file, the uncertainty file should not include units. If the concentration file contains a row of units, the uncertainty file will thus have one less row than the concentration file. Negative values and zero are not permitted as uncertainties. The equation-based uncertainty file provides species-specific parameters that EPA PMF 3.0 uses to calculate the uncertainties for each sample. This file should have one column for each species, with species names as the column header. The first row under the species name is the detection limit, and the second row is the error fraction. The error fraction should be the percentage uncertainty divided by 100. Zeroes or negatives are not permitted for either the detection limit or the percentage uncertainty.

For the sampled concentrations below the MDL (method detection limit), the concentrations are replaced by one half of the MDL, and the missing values are replaced by the arithmetic mean. For concentrations above the MDL, the uncertainty is calculated as the sum of 1/3 of the MDL, analytical uncertainty, and $C2 \times$ concentration (where C2 is the optimal percentage identified using trial and error of PMF runs to assess the appropriate uncertainty weightings and obtain explainable factor profiles). If the concentration is less than or equal to the method detection limit (MDL), the uncertainty is calculated as the sum of $(5/6) \times$ MDL and analytical uncertainty, and four times the arithmetic mean is used as the uncertainty for the missing values (Polissar *et al.*, 1998).

The speciation dataset acquired from the sampling station in this study consisted of 30 species. Categorization of quality of data was based on the signal to noise ratio (S/N) and the percentage of samples above MDL. However some of the species were below the detection limit for most of the samples. Those species which have $S/N \geq 2$ were categorized as having strong data quality, while those with S/N between 0.2 to 2 were categorized as having weak data quality. These species are not likely to provide enough variability in concentration, and therefore contribute to the noise in the results. Those species with S/N ratio below 0.2 were classified as bad values, and were thus excluded from further analysis.

Conditional Probability Function (CPF)

To analyze point the source impacts from various wind directions, the conditional probability function (CPF) was calculated with the source contribution estimated from PMF, coupled with wind direction values measured at the station (Kim *et al.*, 2003a, b; Begum *et al.*, 2004). To minimize the effect of atmospheric dilution, the daily fractional mass contribution from each source relative to the total of all sources was used rather than the absolute source contributions. Here, the fractional contribution from each source is the same as the daily fractional contribution of sources. The same value of the daily fractional mass contribution was assigned to each 1-hr period of a given day to match the 1-hr average wind direction. Specifically, the CPF is defined as:

$$CPF = \frac{m_{\Delta\theta}}{n_{\Delta\theta}} \quad (4)$$

where $m_{\Delta\theta}$ indicates the wind frequency blowing from the direction of $\Delta\theta$ in days with the concentration higher than threshold criterion, while $n_{\Delta\theta}$ indicates the frequency of wind blowing from the direction $\Delta\theta$ in the overall data. In this study, 15 sectors were used ($\Delta\theta = 24^\circ$), while the threshold was set at the upper 25th percentile to define the directionality of the sources. This followed the practice of Kim *et al.* (2004), who conducted tests with several different percentile values of the fractional contribution from each source, and the threshold criterion of upper 25th percentile values was found to have the clearest directionality. Calm wind conditions with a wind speed less than 1 m/s were excluded from this calculation due to isotropic behavior of wind vanes under calm winds. It is expected that the sources are likely to be located in the directions that have high conditional probability values.

RESULTS AND DISCUSSIONS

Elemental Concentrations

The elemental concentrations of PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ are shown in Fig. 2, in which elements are divided into three groups: crustal aerosols, anthropogenic aerosols and ionic compounds (water soluble aerosols). The mean concentrations of metal elements in PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ are 3.18 ± 1.29 , 1.12 ± 0.49 and $0.03 \pm 0.02 \mu\text{g}/\text{m}^3$, respectively, and those of ionic compounds are 9.88 ± 4.52 , 3.13 ± 2.98 and $0.42 \pm 0.33 \mu\text{g}/\text{m}^3$, respectively. The percentages of elements in PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ for the station are 8.23 ± 2.79 , 5.13 ± 2.04 and 2.01 ± 1.27 %, respectively, and those for ionic compounds are 26.30 ± 11.93 , 15.91 ± 9.41 , 30.48 ± 16.20 %, respectively. High percentages of ionic elements are noted in case of the ionic elements for all PM mass concentrations compared to the metal ones. The mean concentrations of OM (Organic Matter, $OM = OC \times 1.6$) and EC concentrations in $PM_{2.5}$ are 5.89 ± 3.82 and $1.18 \pm 0.78 \mu\text{g}/\text{m}^3$, respectively, and the percentages of OM and EC to $PM_{2.5}$ are 26.56 ± 16.67 and 5.38 ± 3.63 %, respectively. OM (26%) is the most abundant species for $PM_{2.5}$ at the station. The average concentrations of PM_{10} , $PM_{2.5}$, and $PM_{0.1}$ are 39.45 ± 11.58 , 21.82 ± 7.50 and $1.42 \pm 0.56 \mu\text{g}/\text{m}^3$, respectively, at Shinjung station. Table 1 presents the average concentrations given in terms of the arithmetic mean and S/N ratio for the particle mass PM, the measured elements and ions. A total of 30 species were selected for the PMF analysis with an S/N ratio greater than 0.2.

The EF values of the various species estimated in this study are shown in Fig. 3. Table 2 shows the EF values for trace metals of the PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ mass concentrations. Sb, Cd and Pb has the highest EF, with Sb being the most enriched element for PM_{10} particles followed by Cd, Pb, Sn, As and Mo, whereas in $PM_{2.5}$ and $PM_{0.1}$ particles Cd is the most enriched element. These elements have high EF values, ranging from 100 to 5,000, indicating that they are only weakly related to soil. K, Rb, Ba, Ni, Cu and Zn are in the range of 5 to 100 in the average EF, and are likely to be

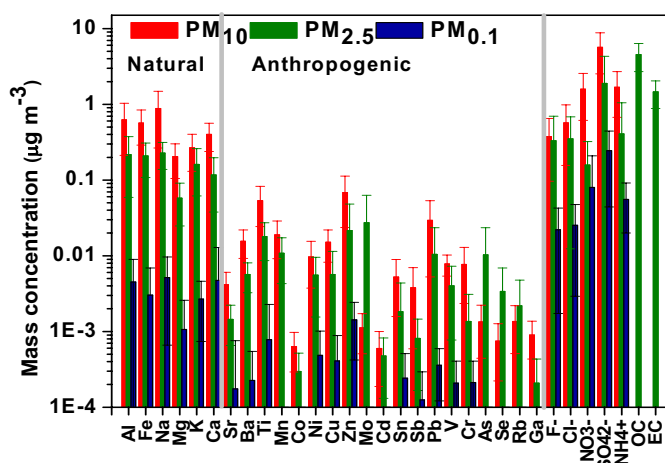


Fig. 2. Concentrations of elements in all PM fractions at Shinjung station, Error bars represent one standard deviation.

Table 1. Summary statistics of PM₁₀, PM_{2.5} and PM_{0.1} species measured at Shinjung station.

	PM ₁₀		PM _{2.5}		PM _{0.1}	
	Avg. (µg/m ³)	S/N	Avg. (µg/m ³)	S/N	Avg. (µg/m ³)	S/N
Al	0.5426	1.0891	0.1717	0.6017	0.0032	0.9836
Fe	0.5405	0.6899	0.1990	0.8812	0.0015	0.9930
Na	0.6560	1.3727	0.2172	0.5787	0.0037	0.9844
Mg	0.2024	0.9214	0.0471	0.6074	0.0005	0.9875
K	0.2226	0.5764	0.1410	0.7350	0.0031	0.9963
Ca	0.3732	0.4744	0.0995	0.8364	0.0014	0.9896
Sr	0.0042	0.9221	0.0013	0.9729	0.0000	0.9966
Ba	0.0147	0.7230	0.0051	0.8972	0.0001	0.9890
Ti	0.0479	0.4749	0.0157	0.7282	0.0004	0.9886
Mn	0.0175	0.9692	0.0098	0.9824	0.0000	0.9983
Co	0.0006	0.9878	0.0003	0.9943	0.0000	0.9996
Ni	0.0084	0.8488	0.0051	0.9126	0.0003	0.9915
Cu	0.0146	0.7593	0.0037	0.9111	0.0004	0.9930
Zn	0.0553	0.7396	0.0143	0.7717	0.0013	0.9827
Mo	0.0010	0.9804	0.0070	0.8015	0.0000	0.9997
Cd	0.0004	0.9883	0.0004	0.9907	0.0000	0.9997
Sn	0.0040	0.9172	0.0005	0.9713	0.0002	0.9953
Sb	0.0031	0.9325	0.0007	0.9852	0.0001	0.9976
Pb	0.0216	0.6319	0.0073	0.8443	0.0004	0.9935
V	0.0078	0.8464	0.0038	0.9224	0.0001	0.9961
Cr	0.0054	0.9415	0.0008	0.9896	0.0002	0.9965
As	0.0012	0.9761	0.0032	0.8471	0.0000	0.9997
Se	0.0006	0.9876	0.0019	0.9452	0.0000	0.9999
Rb	0.0012	0.9732	0.0008	0.9579	0.0000	0.9999
Ga	0.0009	0.9827	0.0002	0.9960	0.0000	0.9998
F	0.3990	2.2186	0.1470	2.5429	0.0189	158.9061
Cl	0.5430	6.7799	0.2260	4.5125	0.0112	269.2728
NO ₃ ⁻	1.3210	16.2280	0.1045	1.5395	0.0203	229.4196
SO ₄ ²⁻	6.3095	81.5235	0.5750	38.1529	0.2170	3.2705
NH ₄ ⁺	1.4980	19.4778	0.0880	7.2201	0.0500	0.5533

affected by both soil and non-soil emission sources. Al, Fe, Ti, Mn, Mg, Co, Ca, V, Sr, Cr and Na have low EF values, ranging from 1 to 5 in the average EF, which indicates that these elements are mostly derived from soil. The results of the PMF analysis confirmed that these elements originate from soil-related emission sources.

Source Apportionment

Identification of a physically meaningful and explainable number of factors is a key step in PMF source apportionment. As suggested by Hopke (2000), and adopted by several recent source apportionment studies (Raman and Hopke, 2007), iterative PMF runs with varying number of factors

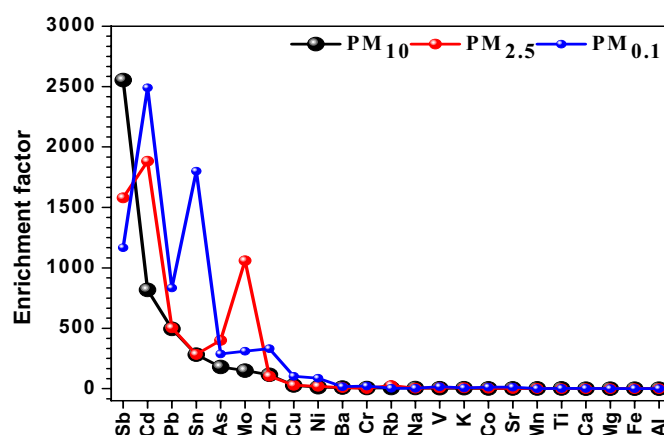


Fig. 3. Enrichment factors of the most abundant trace elements over Shinjung station.

Table 2. Enrichment factors calculated from the trace metal concentration determined in PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ collected in Shinjung.

Metal	Enrichment factors		
	PM_{10}	$PM_{2.5}$	$PM_{0.1}$
Sb	2553.53	1579.62	1167.13
Cd	818.94	1882.94	2490.50
Pb	495.28	504.81	834.50
Sn	282.19	283.33	1799.11
As	180.55	399.70	288.17
Mo	150.36	1059.96	311.27
Zn	115.62	104.38	332.25
Cu	27.15	29.39	101.95
Ni	12.42	20.49	86.34
Ba	8.41	8.79	16.81
Cr	5.60	2.84	21.38
Rb	5.71	26.46	4.27
Na	5.15	3.83	4.18
V	4.59	6.80	16.83
K	4.01	6.94	5.55
Co	2.94	3.96	12.19
Sr	2.16	2.15	12.54
Mn	1.83	2.99	1.31
Ti	1.36	1.31	2.75
Ca	1.02	0.86	1.66
Mg	0.86	0.70	0.62
Fe	1.08	1.15	0.80
Al	1.00	1.00	1.00

and FPEAK were performed. The goodness of model fit parameter 'Q' was evaluated to identify the optimal number of factors, and the optimal solution should lie in this FPEAK range. However, in the current study, a subset of species was used for the analysis, and thus the measured PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ concentrations were included in the PMF runs as an independent variable to obtain mass apportionment without the usual multiple linear regression analysis (Nicolas *et al.*, 2008). The mass fraction distribution of species was used to identify the sources, which included soil dust, vehicle emissions, sea salt, industrial emissions and secondary aerosols. The identified source profiles for PM_{10} ,

$PM_{2.5}$ and $PM_{0.1}$ mass concentrations are shown in Fig. 4, Fig. 5 and Fig. 6, respectively. The source contributions for all concentrations are presented in Fig. 7. The source contribution and source profiles of $PM_{2.5}$ mass concentration with OC, EC concentrations in this station are shown in Fig. 8 and Fig. 9, respectively. Finally the CPF plots are presented in Fig. 10.

PMF analysis was used to study the different source profiles to PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ mass concentrations, and the different source contributions obtained for the sampling period are listed in Table 3. Source contributions were derived from the average of all elements in the matrix obtained by multiplying the rows in factor score and columns in factor loading matrix for the entire dataset, in which the index of rows and columns correspond to the samples for a specified period. From the results of the PMF analysis show that the soil dust (34%), vehicle emissions (33.03%) and secondary aerosols (38.29%) are the major sources for PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ mass concentrations, respectively, at the station.

Source 1: The first source is soil dust, which includes most of the crustal elements and has high concentrations of Fe, Ca, Na, Mg, Al and K. These elements are the major constituents of airborne soil and road dust, and usually make an important contribution to coarse aerosol (Lough *et al.*, 2005). The concentration of Ca is attributed to local construction activities in the area, where concrete was being mixed in the vicinity of the site. Preparation of road surfaces for new pavements also produced an increase in the observed calcium concentrations (Li *et al.*, 2004). PMF analysis showed that 'soil dust' contributed to about 34%, 20.14% and 12.19% of the aerosol mass on average for PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ mass concentrations, respectively, at Shinjung station.

Source 2: The second source is vehicle exhausts, as it is dominated by elements such as Cu, Zn and Sb. Pb and Ni have been widely used as representative markers for vehicle sources. Furusjo *et al.*, (2007) suggested that vehicular emissions are associated with high concentrations of Cu, Zn and Sb. Zn is widely used as a chemical fingerprint for tire wear, while, Cu and Sb are used for break wear. These elements suggest source contributions from motor vehicles,

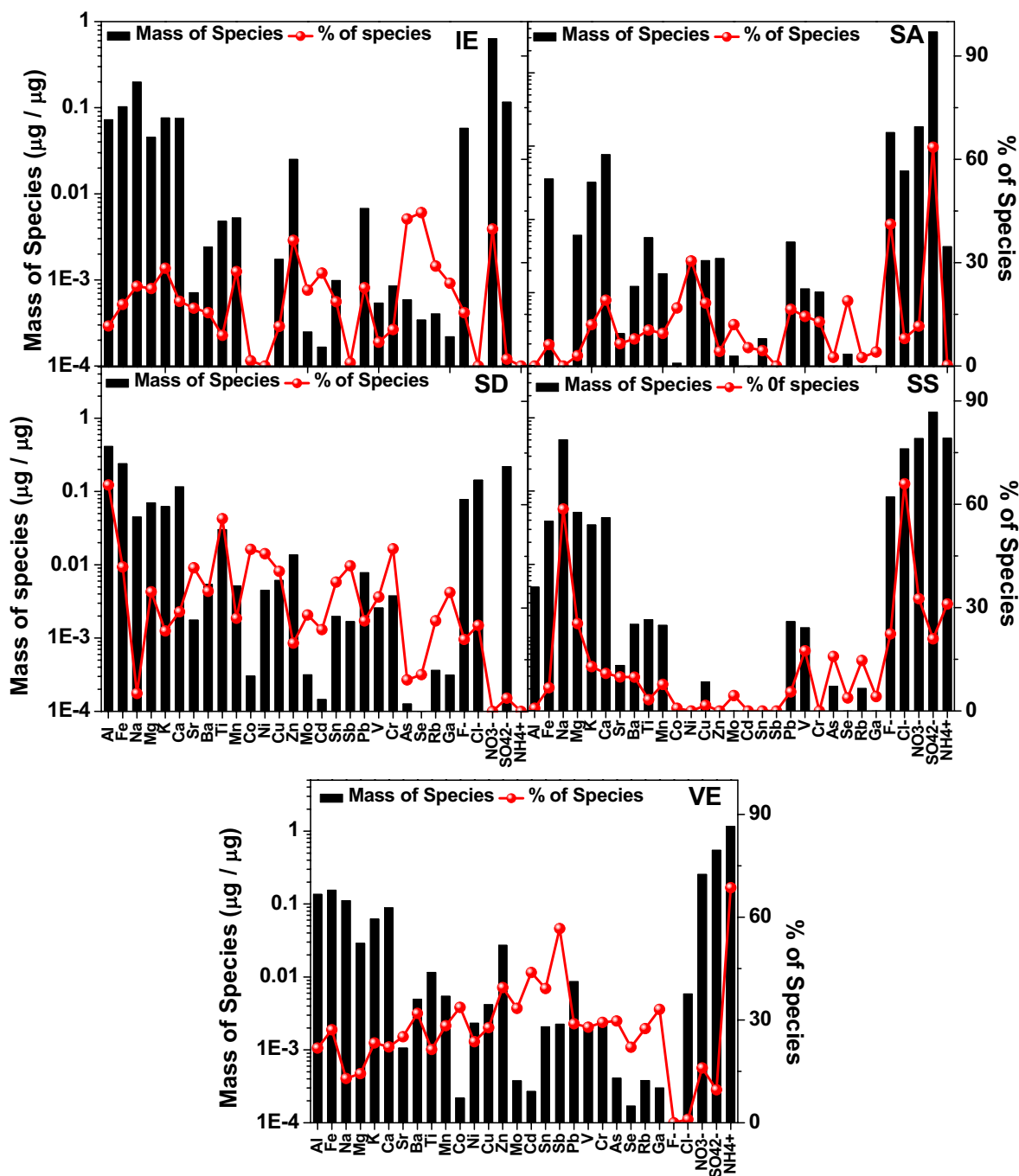


Fig. 4. PMF source profiles of Industrial emissions (IE), secondary aerosols (SA), soil dust (SD), sea salt (SS) and vehicle emissions (VE) in Shinjung station for PM₁₀ mass concentration.

especially those with two-stroke engines, such as motorcycles and motor scooters. In two-stroke engines, fuel and lubricant are mixed and burnt together in the piston chambers, with Zn being emitted. In a four-stroke engine, lubricants are introduced into the cylinders separately, and Zn is emitted from the four-stroke materials (Begum *et al.*, 2005). In our study, this factor was the principal contributor of these metals, and therefore the profile was attributed to vehicle emissions. The results of PMF analysis show that such emissions contributed to about 24.92%, 33.03% and 33.59% of the aerosol mass on average for PM₁₀, PM_{2.5} and PM_{0.1}

mass concentrations, respectively, at Shinjung station.

Source 3: The PMF results show that source 3 had the highest mass fractions of Na and Cl. Source profiles with dominant Na and Cl, as identified in various source apportionment studies conducted in coastal areas have been classified as sea salt (Wu *et al.*, 2007; Guo *et al.*, 2009). Sievering *et al.* (1991) suggested that SO₂ could react on the sea salt particles to produce SO₄²⁻ in addition to the direct reaction of the gas phase H₂SO₄ with NaCl. It appears that this profile includes a combination of sea salt with additional secondary material. The PMF analysis shows

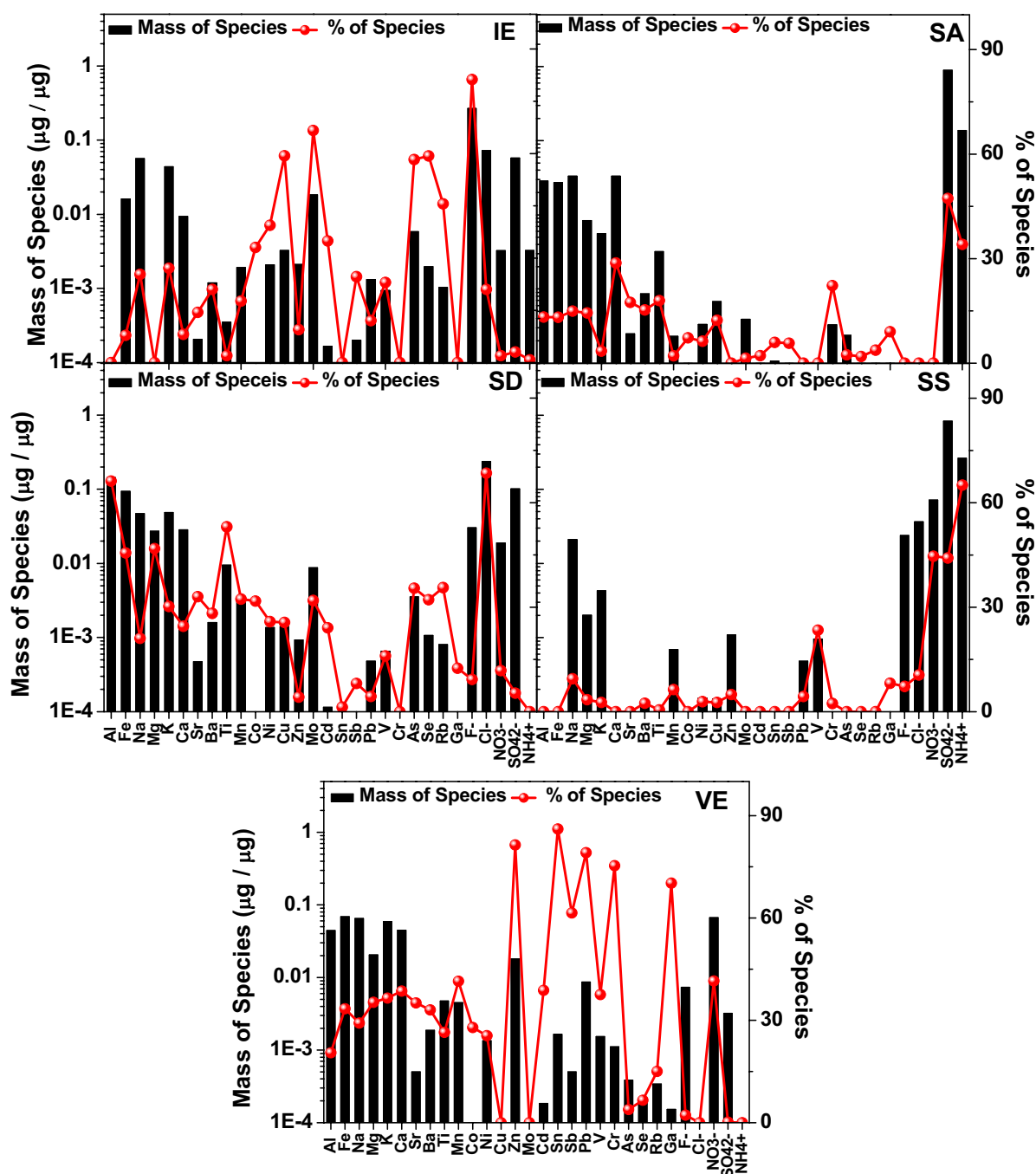


Fig. 5. PMF source profiles of Industrial emissions (IE), secondary aerosols (SA), soil dust (SD), sea salt (SS) and vehicle emissions (VE) in Shinjung station for PM_{2.5} mass concentration.

that sea salt contributed to about 8.4%, 1.12% and 1.16% for PM₁₀, PM_{2.5} and PM_{0.1} mass concentrations, respectively.

Source 4: Secondary aerosol is produced as a result of the reaction and transformation between the emitted pollutants in air, and it is generally difficult to accurately allocate their sources. Secondary aerosols formed by photochemical and other chemical processes are known to be major constituents of the fine particulate matter measured in most industrialized areas. Secondary aerosols of particulate matter include nitrates and sulfates emitted directly from anthropogenic or natural sources, and/or formed in the atmosphere. Biomass

burns, wood burning, and vegetative burning have been characterized as having high concentrations of potassium and sulphates by various source apportionment studies (Ogulei *et al.*, 2006; Wu *et al.*, 2007). This source also reflects regional or long-range transport. Source 4 in the current study was observed to be composed of higher mass fractions of secondary nitrates and sulphates, namely NO₃⁻, NH₄⁺ and SO₄²⁻. In source apportionment studies conducted by Kim *et al.*, (2007) in Ohio, Raman and Hopke (2007) in New York and Tsai and Chen (2006) in southern Taiwan, the source rich in NO₃⁻ and NH₄⁺ was classified as

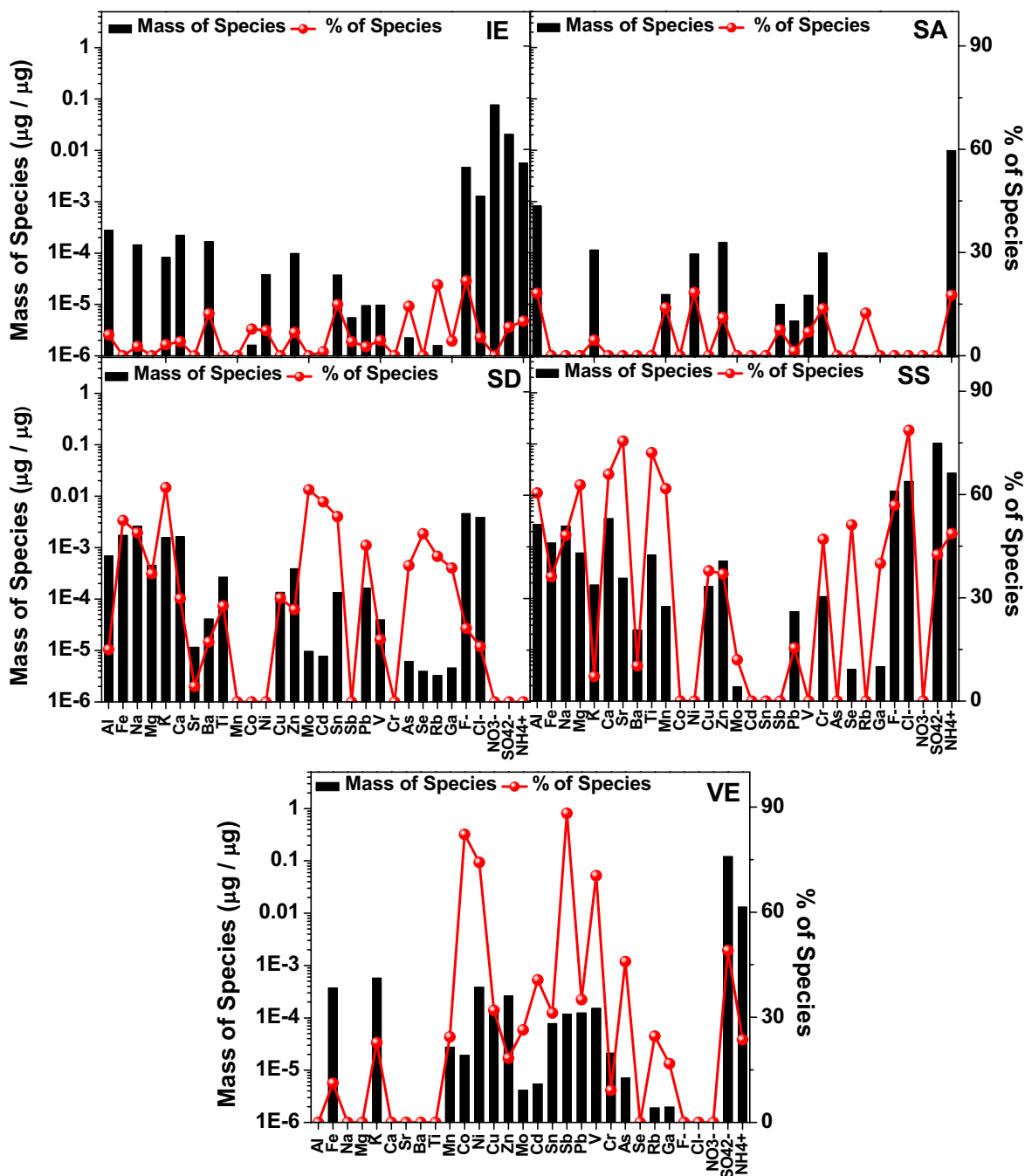


Fig. 6. PMF source profiles of Industrial emissions (IE), secondary aerosols (SA), soil dust (SD), sea salt (SS) and vehicle emissions (VE) in Shinjung station for PM_{0.1} mass concentration.

secondary nitrates, and the secondary sulfates have been identified as a major source of PM_{2.5} in various source apportionment studies (Kim and Hopke, 2004; Ogulei et al., 2006). The current PMF analysis shows that secondary aerosols contributed to about 24.33%, 22.27% and 37.25% for PM₁₀, PM_{2.5} and PM_{0.1} mass concentrations, respectively.

Source 5: Metal elements, such as Cr, Fe, Ni, As and Mn, are present in high concentrations and attributed to industrial sources, as some metal manufacturing plants are located near the sampling sites. The source was estimated as an industrial waste incinerator (made up of several small-

sized incinerators) or Pb-related industries due to the high percentage of Pb as well as the positive contribution of Cl, K, Sb, SO₄²⁻, and NH₄⁺ (Lim et al., 2010). Pb and Sb have been used as markers of incinerator in a number of papers (Lee et al., 2002; Morishita et al., 2006). The results of the PMF analysis show that industrial emissions accounted for about 8.35%, 23.42% and 15.88% for PM₁₀, PM_{2.5} and PM_{0.1} mass concentrations, respectively.

The PMF results given above identified five major sources for three different PM fractions over Shinjung station without OC and EC data. For PM_{2.5}, the five sources

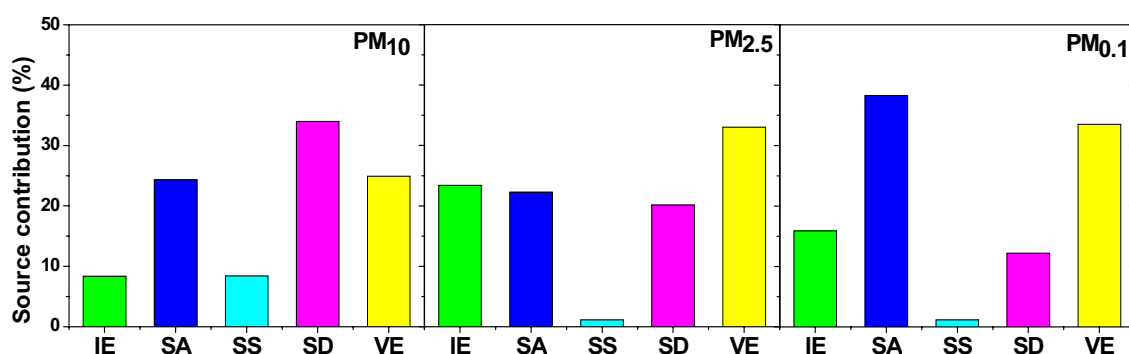


Fig. 7. The source apportionment results from PMF for PM₁₀, PM_{2.5} and PM_{0.1} mass concentrations without OC/EC concentration.

identified from the PMF analysis: soil dust (20.14%), vehicle emissions (33.03%), secondary aerosols (22.27%), industrial emissions (23.42%) and sea salt aerosols (1.12%). Since OC and EC concentration data were available for PM_{2.5} only, additional PMF analysis was conducted and seven source profile categories were identified, as follows: secondary aerosols (17.98%), soil dust (16.84%), vehicle emissions (14.73%), biomass burning (12.70%), industrial emissions (4.70%), sea salt aerosols (3.55%) and fossil fuel burning (29.49%). The source contribution and source profiles are shown in Fig. 8 and Fig. 9. In these figures, high concentrations of Cr, Cd, Ni, Se, V and Zn clearly indicate the source is fossil fuel combustion. Cr and Cd are known to occur at high temperatures during the combustion of coal, oil, refuse, and so on. Ni and V are widely used as markers for the combustion of heating fuel (Vallius *et al.*, 2005), while Se and Zn are representative marker species for oil-fired power plants and coal combustion, respectively (Lee *et al.*, 2002; Morishita *et al.*, 2006). Metal elements and ionic compounds, such as K, Se, Na, Cl, NH₄⁺ and SO₄²⁻ are present in high concentrations and attributed to biomass burning. The results show that OC and EC are contributed by traffic emissions, biomass burning, wood burning and vegetative burning, different to the source contributions for PM_{2.5} obtained without OC and EC data. This suggests the importance of including OC and EC data in future studies of source apportionment by PMF analysis for all PM fractions.

Conditional Probability Function Study

The estimation of the average mass contribution of source factors by wind sectors is based on the values of the wind sector factor loading obtained from the PMF analysis. The results are shown in Fig. 10 for the major source factors, as well as the total contribution from all the source factors. Since the prevailing winds were in the NE sector, the directional contributions of most of the source factors were also dominated by these wind sectors, as expected, while secondary aerosols and soil dust aerosols were directionality contributed in the NE and SW sectors. The vehicle emission factor directionality was contributed in the NE and NW directions due to highways and heavy traffic. The sea salt aerosols directionality were contributed in the directions of the SE and SW sectors, which confirms that sea breeze was the major contributing factor for sea spray.

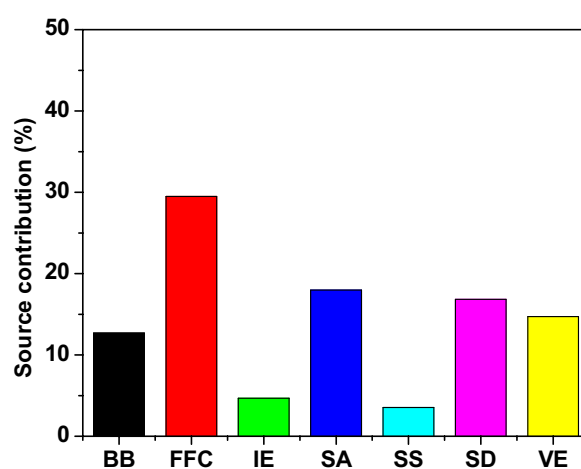


Fig. 8. The source apportionment results from PMF for PM_{2.5} mass concentration with OC/EC mass concentrations.

Comparison of Particle Concentrations and Source Contributions in Asia

Particulate mass concentration data collected in Asia are presented in Table 4. The highest average PM₁₀ concentration was obtained in urban areas like Agra, India (ranged from 270–290 µg/m³) (Pipal *et al.*, 2011) and Guangzhou, China (ranged from 120–130 µg/m³) (Cao *et al.*, 2004). The highest average PM_{2.5} concentration was also obtained in urban areas like Agra, India (ranged from 80–90 µg/m³) (Pipal *et al.*, 2011) and Guangzhou, China (78 µg/m³) (Cao *et al.*, 2004) and Ordos, China (51 µg/m³) (Wang *et al.*, 2012). The PM₁₀, PM_{2.5} and PM_{0.1} concentrations obtained in this study were found to be the lowest in the Asian region, with mean values of about 39.45 ± 11.58, 21.82 ± 7.50 and 1.42 ± 0.56 µg/m³, respectively. The observed average PM₁₀ mass concentrations are much lower than the air quality standard of 125 µg/m³ (daily average) of the Taiwan Environmental Protection Agency (EPA), and PM_{2.5} mass concentrations are also lower than the air quality standards that will be adopted in the future, 35 µg/m³ (daily average).

Table 5 summarizes the average different types of source contribution in this study and some other recent source apportionment studies. It is clear that the number and type of source factors derived from the PMF analysis in this work are similar to those reported in other studies. Most of

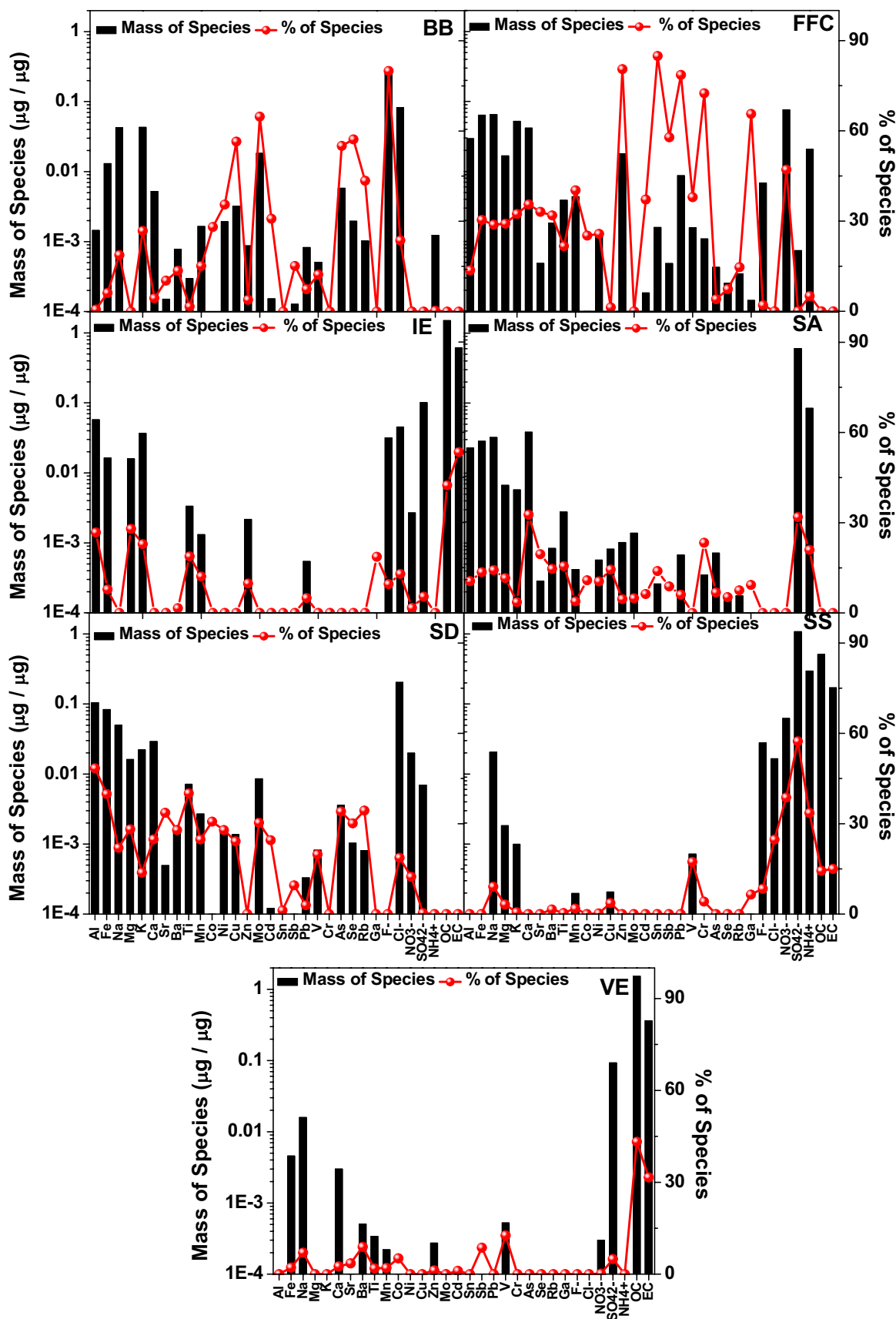


Fig. 9. PMF source profiles of biomass burning (BB), fossil fuel combustion (FFC), Industrial emissions (IE), secondary aerosols (SA), soil dust (SD), sea salt (SS) and vehicle emissions (VE) in Shinjung station for PM_{2.5} mass concentration with OC/EC mass concentration.

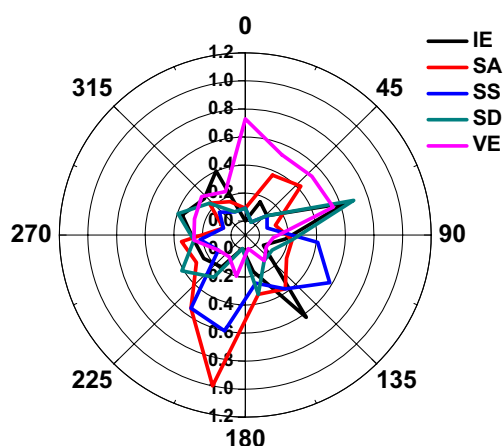


Fig. 10. CPF plot for the source contributions for the source identified at Shinjung.

the earlier researchers used hundreds of samples and more than 10 species in their analysis. In general, three to eight source factors were resolved for PM_{10} particles, and five–eight for $PM_{2.5}$ particles, depending on the site characteristics and sampling periods in the studies. For most of the studies, a similar set of major source types was always identified.

For example, soil dust, vehicle emissions, industrial emissions and secondary aerosols were the most common major source types identified for particulate matters. Using a reasonably sized data set, this study shows the current PMF analysis is also capable of resolving source factors which are similar to those reported in the literature with larger data sets.

CONCLUSIONS

Source apportionment of PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ measured in Shinjung station was conducted using the advanced factor analysis model, PMF3. Soil dust was identified as the major source accounting for 34% of the PM_{10} apportioned mass, followed by vehicle emissions (24.92%), secondary aerosols (24.33%), sea salt (8.4%) and industrial emissions (8.35%). In the case of $PM_{2.5}$, vehicle emissions were identified as the major source (33.03%), while sea salt (1.12%) was identified as the least importance one. However, in the case of $PM_{0.1}$, secondary aerosols (38.29%) were identified as major source and sea salt (1.16%) as the least importance one. From the CPF analysis, the results of directional dependencies matched well with the location of known sources in the area. The results clearly showed that the NE wind sectors were dominant for most of the source

Table 3. Source contributions percentage derived from PMF model for the sampling period in Shinjung station.

Source	PM_{10} mass (%)	$PM_{2.5}$ mass (%)	$PM_{0.1}$ mass (%)
Industrial emissions	8.35	23.42	15.88
Secondary aerosols	24.33	22.27	37.25
Soil dust	34.0	20.14	12.19
Sea salt	8.4	1.12	1.15
Vehicle emissions	24.92	33.03	33.59

Table 4. PM_{10} and $PM_{2.5}$ mass concentrations in different stations in Asia.

Stations	Year	PM_{10} Concentration ($\mu\text{g}/\text{m}^3$)	$PM_{2.5}$ Concentration ($\mu\text{g}/\text{m}^3$)	References
Shinjung, Taiwan	2011	39.45	21.82	Present study
Ordos, China	2005	89.12	51.81	Wang <i>et al.</i> (2012)
Guangzhou, China	2002	124.7	78.1	Cao <i>et al.</i> (2004)
Hong Kong, China	2002	41.4	31	Cao <i>et al.</i> (2004)
Shenzhen, China	2002	75.1	47.1	Cao <i>et al.</i> (2004)
Tianjin, China	2005	59.2	47.4	Wang <i>et al.</i> (2008)
Haarlemmerweg, Netherlands	2008	27.5	17.8	Boogaard <i>et al.</i> (2011)
Agra, India (Road side)	Apr. 2010–June 2010	278	90	Pipal <i>et al.</i> (2011)
Agra, India (Semi rural site)	Apr. 2010–June 2010	234	89	Pipal <i>et al.</i> (2011)
Seoul, Korea		50.5	--	Kim <i>et al.</i> (2003)
Taejon, Korea		92.2	--	Kim <i>et al.</i> (2002)
Taiwan	2005	59.2	47.4	Wang <i>et al.</i> (2008)
Guanyin, Taiwan	2000–08	52.67	28.0	Fang <i>et al.</i> (2010)
Banciao, Taiwan	2000–08	52.22	30.7	Fang <i>et al.</i> (2010)
Shalu, Taiwan	2000–08	56.22	31.3	Fang <i>et al.</i> (2010)
Erlin, Taiwan	2000–08	66.67	35.7	Fang <i>et al.</i> (2010)
Tainan, Taiwan	2000–08	70.89	42.7	Fang <i>et al.</i> (2010)
Hualen, Taiwan	2000–08	34.44	20.7	Fang <i>et al.</i> (2010)
Taitung, Taiwan	2000–08	35.33	15.7	Fang <i>et al.</i> (2010)
Lahore, Pakistan	2006	459	--	Zhang <i>et al.</i> (2008)

Table 5. Average contribution (%) from major source types, compiled from the results reported in recent source apportionment studies.

Study	No. of samples	Avg. concen. ($\mu\text{g}/\text{m}^3$)	No. of factors	Sea salt (%)	Crustal/Soil dust (%)	Vehicle emissions (%)	Secondary aerosols (%)	Other combustion/Industrial emissions (%)	Reference
Shinjung, Taiwan, 2011	18 PM ₁₀	39.45	5	8.4	34.0	24.92	24.33	8.35	Present study
	18 PM _{2.5}	21.82	5	1.12	20.14	33.03	22.27	23.42	
	18 PM _{0.1}	1.42	5	1.15	12.19	33.59	37.25	15.88	
Salamanca, Mexico	140 PM _{2.5}	45	6	--	19.9	13.8	11.7	13.6	Murillo et al. (2012)
	135 PM ₁₀	89.12	5	--	39.87	30.16	14.42	1.82	Wang et al. (2012)
Ordos, China	122 PM _{2.5}	51.81	4	--	35.81	22.67	32.35	4.57	Zeng et al., (2010)
	14 PM ₁₀	305	8	---	12	13	16	30	
Taiyuan, China Apr. 2001–Jan. 02	CMB analysis								Lestari and Mauliadi (2009)
Bandung, Indonesia, 2001–2007	180 coarse 180 fine	19.0 48.0	5 7	23 13	19 ---	-- 22	18 25	40 40	
Athens, Greece, 3 sites, 2002	62 coarse 67 fine	18.0 41.0	3 5	20 24	30 18	50 34	-- --	-- 24	Karaniasiou et al. (2009)
	243 coarse 279 fine	12.6 27.7	8 8	29 3	67 14	28 29	3 45	2 10	
Barcelona, Spain, 2003–2007									Amato et al. (2009)
Chicago, Illinois, US, 2001–2003	372 fine	15.5	10	3	6	23	58	12	Rizzo and Scheff (2007)
Southwestern Oregon, US, 2000–2004	493 fine	3.2	9	10	9	3	40	38	Hwang and Hopke (2007)
Midwest US, 5 Cities, 2002–2005	1207 fine	14.5	7–9	--	5	30	51	14	Buzcu-Guven et al. (2007)
Eagle Farm, Brisbane, Australia, June 2001	28 coarse 28 fine	10.2 7.2	3 7	2 58	40 8	32 30	-- 22	-- 32	Chan et al. (2011)
	437 coarse 437 fine	9.8 5.9	4 8	38 20	38 6	5 24	19 25	-- 22	
4 Australian Cities, 8 sites, 2003–2004									Chan et al. (2008b)

factors observed, except in the case of industrial emissions and sea salt aerosols. The results of this study can be applied to reasonably sized data sets to enable source location and apportionment of PM in local pollution problems. This study can also help stakeholders and policymakers in understanding the influence of regional and local sources of PM₁₀, PM_{2.5} and PM_{0.1} on urban areas, and thus to identify effective emission control strategies.

ACKNOWLEDGEMENTS

The authors would like to express their gratitude to the Taiwan EPA project (EPA-100-1602-02-01) and Taiwan NSC project (NSC 98-2221-E-009-020-MY3) for financial support.

REFERENCES

- Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Perez, N. and Hopke, P.K. (2009). Quantifying Road Dust Resuspension in Urban Environment by Multilinear Engine: A Comparison with PMF2. *Atmos. Environ.* 43: 2770–2780.
- Atkinson, R.W., Fuller, G.W., Anderson, H.R., Harrison, R.M. and Armstrong, B. (2010). Urban Ambient Particle Metrics and Health: A Time-Series Analysis. *Epidemiology* 21: 501–511.
- Barmpadimos, I., Nufer, M., Oderbolz, D.C., Keller, J., Aksoyoglu, S., Hueglin, C., Baltensperger, and Prévôt, A.S.H. (2011). The Weekly Cycle of Ambient Concentrations and Traffic Emissions of Coarse (PM₁₀-PM_{2.5}) Atmospheric Particles. *Atmos. Environ.* 45: 4580–4590.
- Begum, B.A., Kim, E., Biswas, S.K. and Hopke, P.K., (2004). Investigation of Sources of Atmospheric Aerosol at Urban and Semi-urban Areas in Bangladesh. *Atmos. Environ.* 38: 3025–3038.
- Begum, B.A., Biswas, S.K., Kim, E., Hopke, P.K. and Khaliqzaman, M. (2005). Investigation of Sources of Atmospheric Aerosol at a Hot Spot Area in Dhaka, Bangladesh. *J. Air Waste Manage. Assoc.* 55: 227–240.
- Boogaard, H., Kos, G.P.A., Weijers, E.P., Janssen, N.A.H., Fischer, P.H., van der Zee, S.C., de Hartog, S.C. and Hoek, G. (2011). Contrast in Air Pollution Components between Major Streets and Background Locations: Particulate Matter Mass, Black Carbon, Elemental Composition, Nitrogen Oxide and Ultrafine Particle Number. *Atmos. Environ.* 45: 650–658.
- Buzcu-Guven, B., Brown, S.G., Frankel, A., Hafner, H.R. and Roberts, P.T. (2007). Analysis and Apportionment of Organic Carbon and Fine Particulate Matter Sources at Multiple Sites in the Midwestern United States. *J. Air Waste Manage. Assoc.* 57: 606–619.
- Cao, J.J., Lee, S.C., Ho, K.F., Zou, S.C., Fung, K., Li, Y., Watson, J.G. and Chow, J.C. (2004). Spatial and Seasonal Variations of Atmospheric Organic Carbon and Elemental Carbon in Pearl River Delta Region, China. *Atmos. Environ.* 38: 4447–4456.
- Chan, Y.C., Christensen, E., Golding, G., King, G., Gore, W., Cohen, D.D., Hawas, O., Stelcer, E., Simpson, R., Denison, L. and Wong, N. (2008). Source Apportionment of Ambient Volatile Organic Compounds in Major Cities in Australia by Positive Matrix Factorisation. *Clean Air Environ. Qual.* 42: 22–29.
- Chan, Y.C., Hawas, O., Hawker, D., Vowles, P., Cohen, D.D., Stelcer, E., Simpson, R., Golding, G. and Christensen, E. (2011). Using Multiple Type Composition Data and Wind Data in PMF Analysis to Apportion and Locate Sources of Air Pollutants. *Atmos. Environ.* 45: 439–449.
- Chang, S.C., Chou, C.C.K., Chan, C.C. and Lee, C.T. (2010). Temporal Characteristics from Continuous Measurements of PM_{2.5} and Speciation at the Taipei Aerosol Supersite from 2002 to 2008. *Atmos. Environ.* 44: 1088–1096.
- Chen, S.C., Tsai, C.J., Huang, C.Y., Chen, H.D., Chen, S.J., Lin, C.C., Tsai, J.H., Chou, C.C.K., Lung, S.C.C., Hang, W.R., Roam, G.D., Wu, W.Y., Smolik, J. and Dzumbova, L. (2010). Chemical Mass Closure and Chemical Characteristics of Ambient Ultrafine Particles and other PM Fractions. *Aerosol Sci. Technol.* 44: 713–723.
- Choosong, T., Chomanee, J., Tekasakul, P., Tekasakul, S., Otani, Y., Hata, M. and Furuuchi, M. (2010). Workplace Environment and Personal Exposure of PM and PAHs to Workers in Natural Rubber Sheet Factories Contaminated by Wood Burning Smoke. *Aerosol Air Qual. Res.* 10: 8–21.
- Fang, G.C. and Chang, S.C. (2010). Atmospheric Particulate (PM₁₀ and PM_{2.5}) Mass Concentration and Seasonal Variation Study in the Taiwan Area during 2000–2008. *Atmos. Res.* 98: 368–377.
- Furusjo, E., Sternbeck, J. and Cousins, A.P. (2007). PM₁₀ Source Characterization at Urban and Highway Roadside Locations. *Sci. Total Environ.* 387: 206–219.
- Guo, H., Ding, A.J., So, K.L., Ayoko, G., Li, Y.S. and Hung, W.T. (2009). Receptor Modeling of Source Apportionment of Hong Kong Aerosols and the Implication of Urban and Regional Contribution. *Atmos. Environ.* 43: 1159–1169.
- Hopke, P.K. (1991). *Receptor Modeling for Air Quality Management*, Elsevier, Amsterdam.
- Hopke, P.K. (2000). A Guide to Positive Matrix Factorization EPA Workshop Proceedings Materials from the Workshop on UNMIX and PMF as Applied to PM_{2.5}.
- Hwang, I. and Hopke, P.K. (2007). Estimation of Source Apportionment and Potential Source Locations of PM_{2.5} at a West Coastal IMPROVE Site. *Atmos. Environ.* 41: 506–518.
- IPPC (2007). Climate Change 2007: the Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the IPCC.
- Karanasiou, A.A., Siskos, P.A. and Eleftheriadis, K. (2009). Assessment of Source Apportionment by Positive Matrix Factorization Analysis on Fine and Coarse Urban Aerosol Size Fractions. *Atmos. Environ.* 43: 3385–3395.
- Khan, M.F., Hirano, K. and Masunaga, S. (2010). Quantifying the Sources of Hazardous Elements of Suspended Particulate Matter Aerosol Collected in Yokohama, Japan. *Atmos. Environ.* 44: 2646–2657.

- Kim, E., Hopke, P.K. and Edgerton, E. (2003a). Source Identification of Atlanta Aerosol by Positive Matrix Factorization. *J. Air Waste Manage. Assoc.* 53: 731–739.
- Kim, E., Larson, T.V., Hopke, P.K., Slaughter, C., Sheppard, L.E. and Claiborne, C. (2003b). Source Identification of PM_{2.5} in an Arid Northwest U.S. City by Positive Matrix Factorization. *Atmos. Res.* 66: 291–305.
- Kim, E. and Hopke, P.K. (2004). Source Apportionment of Fine Particles in Washington, DC, Utilizing Temperature-resolved Carbon Fractions. *J. Air Waste Manage. Assoc.* 53: 773–785.
- Kim, M., Deshpande, S.R. and Crist, K.C. (2007). Source Apportionment of Fine Particulate Matter (PM_{2.5}) at a Rural Ohio River Valley Site. *Atmos. Environ.* 41: 9231–9243.
- Kim, N.K., Kim, Y.P. and Kang, C.H. (2011). Long-term Trend of Aerosol Composition and Direct Radiative Forcing due to Aerosols over Gosan: TSP, PM₁₀, and PM_{2.5} Data between 1992 and 2008. *Atmos. Environ.* 45: 6107–6115.
- Lee, J.H., Yoshida, Y., Turpin, B.J., Hopke, P.K., Poirot, P.J., Liroy, P.J. and Oxley, J.C. (2002). Identification of Sources Contributing to Mid-Atlantic Regional Aerosol. *J. Air Waste Manage. Assoc.* 52: 1186–1205.
- Lee, J.H. and Hopke, P.K. (2006). Apportioning Sources of PM_{2.5} in St. Luis, MO Using Speciation Trends Network Data. *Atmos. Environ.* 40: S360–S377.
- Lestari, P. and Mauliadi, Y.D. (2009). Source Apportionment of Particulate Matter at Urban Mixed Sites in Indonesia Using PMF. *Atmos. Environ.* 43: 1760–1770.
- Li, Z., Hopke, P.K., Husain, L., Qureshi, S., Dutkiewicz, V.A., Schwab, J.J., Drewnick, F. and Demerjian, K.L. (2004). Sources of Fine Particle Composition in New York City. *Atmos. Environ.* 38: 6521–6529.
- Lim, J.M., Lee, J.H., Moon, J.H., Chung, Y.S. and Kim, K.H. (2010). Source Apportionment of PM₁₀ at a Small Industrial Area Using Positive Matrix Factorization. *Atmos. Res.* 95: 88–100.
- Lough, G.C., Schauer, J.J., Park, J.S., Shafer, M.M., DeMinter, J.T. and Weinstein, J.P. (2005). Emissions of Metals Associated with Motor Vehicle Roadways. *Environ. Sci. Technol.* 39: 826–836.
- Marple, V.A., Rubow, K.L. and Behm, S.M. (1991). A Micro Orifice Uniform Deposit Impactor (MOUDI): Description, Calibration, and Use. *Aerosol Sci. Technol.* 14: 434–446.
- Mcbride, S.J., Norris, G.A., Williams, R.W. and Neas, L.M. (2011). Bayesian Hierarchical Modeling of Cardiac Response to Particulate Matter Exposure. *J. Exposure Sci. Environ. Epidemiol.* 21: 74–91.
- Morishita, M., Keeler, G.J., Wagner, J.G. and Harkema, J.R. (2006). Source Identification of Ambient PM_{2.5} during Summer Inhalation Exposure Studies in Detroit, MI. *Atmos. Environ.* 40: 3823–3834.
- Murillo, J.H., Ramos, A.C., Carcia, F.A., Jimenez, S.B., Cardenas, B. and Mizohata, A. (2012). Chemical Composition of PM_{2.5} Particles in Salamanca, Guanajuato Mexico: Source Apportionment with Receptor Models. *Atmos. Res.* 107: 31–41.
- Nicolas, J., Chiari, M., Crespo, J., Orellana, I.G., Lucarelli, F., Nava, S., Pastor, C. and Yubero, E. (2008). Quantification of Saharan and Local Dust Impact in an Arid Mediterranean Area by the Positive Matrix Factorization (PMF) Technique. *Atmos. Environ.* 42: 8872–8882.
- Ning, Z. and Sioutas, C. (2010). Atmospheric Processes Influencing Aerosols Generated by Combustion and the Influence of their Impact on Public Exposure: A Review. *Aerosol Air Qual. Res.* 10: 43–58.
- Ogulei, D., Hopke, P.K., Zhou, L., Pancras, J.P., Nair, N. and Ondov, J.M. (2006). Source Apportionment of Baltimore Aerosol from Combined Size Distribution and Chemical Composition Data. *Atmos. Environ.* 40: 396–410.
- Paatero, P. and Tapper, U. (1993). Analysis of Different Modes of Factor Analysis as Least Squares Fit Problem. *Chemom. Intell. Lab. Syst.* 18: 183–194.
- Paatero, P. and Tapper, U. (1994). Positive Matrix Factorization: A Non-negative Factor Model with Optimal Utilization of Error Estimates of Data Values. *Environmetrics* 5: 111–126.
- Paatero, P. (1997). Least Squares Formulation of Robust Nonnegative Factor Analysis. *Atmos. Environ.* 37: 23–35.
- Pipal, A.S., kulshrestha, A. and Taneja, A. (2011). Characterization and Morphological Analysis of Airborne PM_{2.5} and PM₁₀ in Agra Located in North Central India. *Atmos. Environ.* 45: 3621–3630.
- Polissar, A.V. (1998). Atmospheric Aerosol over Alaska: 2. Elemental Composition and Sources. *J. Geophys. Res.* 10: 19045–19057.
- Raman, R.S. and Hopke, P.K. (2007). Source Apportionment of Fine Particles Utilizing Partially Speciated Carbonaceous Aerosol Data at Two Rural Locations in New York State. *Atmos. Environ.* 41: 7923–7939.
- Rizzo, M. and Scheff, P.A. (2007). Fine Particulate Source Apportionment Using Data from the USEOA Speciation Trends Network in Chicago, Illinois: Comparison of Two Source Apportionment Models. *Atmos. Environ.* 41: 6276–6288.
- Sahu, S.K., Beig, G. and Parkhi N.S. (2011). Emissions Inventory of Anthropogenic PM_{2.5} and PM₁₀ in Delhi during Commonwealth Games 2010. *Atmos. Environ.* 45: 6180–6190.
- Sievering, H., Boatman, J., Galloway, J., Keene, W., Kim, Y., Luria, M. and Ray, J. (1991). Heterogeneous Sulfur Conversion in Sea-salt Aerosol Particles: The Role of Aerosol Water Content and Size Distribution. *Atmos. Environ.* 25: 1479–1487.
- Taylor, S.R. and McLennan, S.H. (1985). *The Continental Crust: Its Composition and Evolution*, Blackwell, Oxford.
- Tsai, Y.I. and Chen, C.L. (2006). Atmospheric Aerosol Composition and Source Apportionments to Aerosol in Southern Taiwan. *Atmos. Environ.* 40: 4751–4763.
- USEPA (2008). EPA Positive Matrix Factorization (PMF) 3.0 Fundamentals and User Guide, USEPA Office of Research and Development.
- Vallius, M., Janssen, N.A.H., Heirich, J., Hoek, G., Ruuskanen, J., Cyrys, J., Grieken, R.V., Hartog, J.J., Kreyling, W.G. and Pekkanen, J. (2005). Sources and

- Elemental Composition of Ambient PM_{2.5} in Three European Cities. *Sci. Total Environ.* 337: 147–162.
- Wang Z.S., Wu, Ting., Shi, G.L., Fu, X., Tian, Y., Feng, Y.C., Wu, X.F., Wu, G., Bai, X.P. and Zhang, W.J. (2012). Potential Source Analysis for PM₁₀ and PM_{2.5} in Autumn in a Northern City in China. *Aerosol Air Qual. Res.* 12: 39–48.
- Wang, W.C., Chen, K.S., Chen, S.J., Lin, C.C., Tsai, J.H., Lai, C.H. and Wang, S.K. (2008). Characteristics and Receptor Modeling of Atmospheric PM_{2.5} at Urban and Rural Sites in Pingtung, Taiwan. *Aerosol Air Qual. Res.* 8: 112–129.
- Wu, C.F., Larson, T.V., Wu, S.Y., Williamson, J., Westberg, H.H. and Liu, L.J.S. (2007). Source Apportionment of PM_{2.5} and Selected Hazardous Air Pollutants in Seattle. *Sci. Total Environ.* 386: 42–52.
- Zeng, F., Shi, G.L., Li, X., Feng, Y.C., Bi, X.H., Wu, J.H. and Xue, Y.H. (2010). Application of a Combined Model to Study the Source Apportionment of PM₁₀ in Taiyuan, China. *Aerosol Air Qual. Res.* 10: 177–184.
- Zhang, Y.X., Quraishi, T. and Schauer, J.J. (2008). Daily Variations in Sources of Carbonaceous Aerosol in Lahore, Pakistan during a High Pollution Spring Episode. *Aerosol Air Qual. Res.* 8: 130–146.

Received for review, April 12, 2012

Accepted, May 3, 2012