



## Characteristics of Concentrations and Metal Compositions for PM<sub>2.5</sub> and PM<sub>2.5-10</sub> in Yunlin County, Taiwan during Air Quality Deterioration

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### ABSTRACT

Concentrations of twenty trace metals in ambient fine and coarse particles were characterized during the period of air quality deterioration of elevated particulate matters (PM episode) in winter in the suburban area, Yunlin County, Taiwan. The potential emission sources of fine- and coarse-size metals were identified using the enrichment factor (EF) analysis, principal component analysis (PCA) and backward trajectory model. The high concentrations of fine and coarse particles and their associated metals were observed during the PM episode in Yunlin County. Al, Fe, and Ca were predominant in fine and coarse particles for sampling periods, accounting for 75–85% of all selected elements. When the PM episode occurred, mean concentrations of fine-size Pb and As were increased to 132% and 116%, respectively, while average concentrations of coarse-size Fe, Al, Mn, Ti, and Co were increased to 1.1–2.2 times, in comparison with non-episode period. The mean value of fine-size As (6.67 ng m<sup>-3</sup>) obtained from the episode period exceeds the proposed European Union standard (6 ng m<sup>-3</sup>). EF values were decreased with increasing particle size for Sb, Se, As, Mo, Cr, Cu, V, Pb, Zn, Ni and Cd in both episode periods. Fine-size metals is likely to refer to coal combustion (36.5%), iron and steel industry (30.5%), vehicular emission and oil burning (16.3%), and Cu smelting source (13.1%), while coarse-size metals are associated with soil dust and crustal elements (61.6%), traffic-related re-suspended road dust (19.4%), industrial process (10.6%) and coal/oil combustion (6.3%) for the PM episode.

**Keywords:** Trace metals; Episode; Enrichment factor; Source apportionment.

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### INTRODUCTION

Atmospheric particulate matters (PM) can affect the visibility of the atmosphere and play key roles in the formation of acid rain and climate change and in regional air quality deterioration (Appel *et al.*, 1985; Tai *et al.*, 2010; Cheng *et al.*, 2012). Air quality deterioration of elevated PM associated with the proliferation of respiratory and cardiovascular diseases have been reported (Peters *et al.*, 1997; Peters *et al.*, 2000; Calverley, 2003; Trianti *et al.*,

2014). Besides, elevated PM mass concentrations, trace metals in ambient PMs have been recognized as an important risk factor of health in human and animal studies (Chen and Nadziejko 2005; Chen and Lippmann 2009; Bell *et al.*, 2014; Gehring *et al.*, 2015). In order to initiate effective control strategies for ambient PM and metallic elements, their major sources and contributions from each source need to be investigated thoroughly.

In general, a bimodal distribution of ambient PM has been reported for many sites (Aceves and Grimalt 1993; Horvath *et al.*, 1996; Hering *et al.*, 1997). The coarse particle (the aerodynamic diameter of larger than 2.5 μm and smaller than 10 μm, referring PM<sub>2.5-10</sub>) is mainly from crustal materials, paved-road dust, non-catalyst equipped gasoline engines and background sea salt. The fine particle (the aerodynamic diameter of less than 2.5 μm, referring PM<sub>2.5</sub>) is emitted mostly from anthropogenic rather than natural sources or formed by vapour nucleation/condensation mechanisms. Knowledge of sources and their contributions to coarse and

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fine particles is crucial in making feasible policies for their levels to improve ambient air quality and protect human health.

Source apportionment methods for fine and/or coarse particles have been widely performed around the world (Manoli *et al.*, 2002; Mysliwiec and Kleeman 2002; Viana *et al.*, 2008; Stortini *et al.*, 2009; Li *et al.*, 2013; Kaul *et al.*, 2014; Tao *et al.*, 2014). Such techniques have also been increasingly applied for the past decade in the urban cities and areas, Taiwan (Wang *et al.*, 1998; Yuan *et al.*, 2000; Chang *et al.*, 2006; Liang *et al.*, 2013). In addition to the large amount of ionic components, carbon and major crustal elements, the composition and characterization of trace metals in fine and coarse particles can provide important information on the sources, while these trace elements only contain a small proportion of the total PM mass.

To date, ambient PM pollution is still one of the most concerned environmental issues in Taiwan. More frequent episodes of air quality deterioration due to elevated PM have occurred in particular locations (western coastal central Taiwan) (Kuo *et al.*, 2010, 2013) and during particular seasons (winter) in Taiwan (Lin *et al.*, 2004; Tsai *et al.*, 2013). Kuo *et al.* (2010) have reported that trace metals with a high level in fine and coarse particles could be observed in Yunlin County of central Taiwan due to a river dust episode in winter. In this study, source identification methods are used

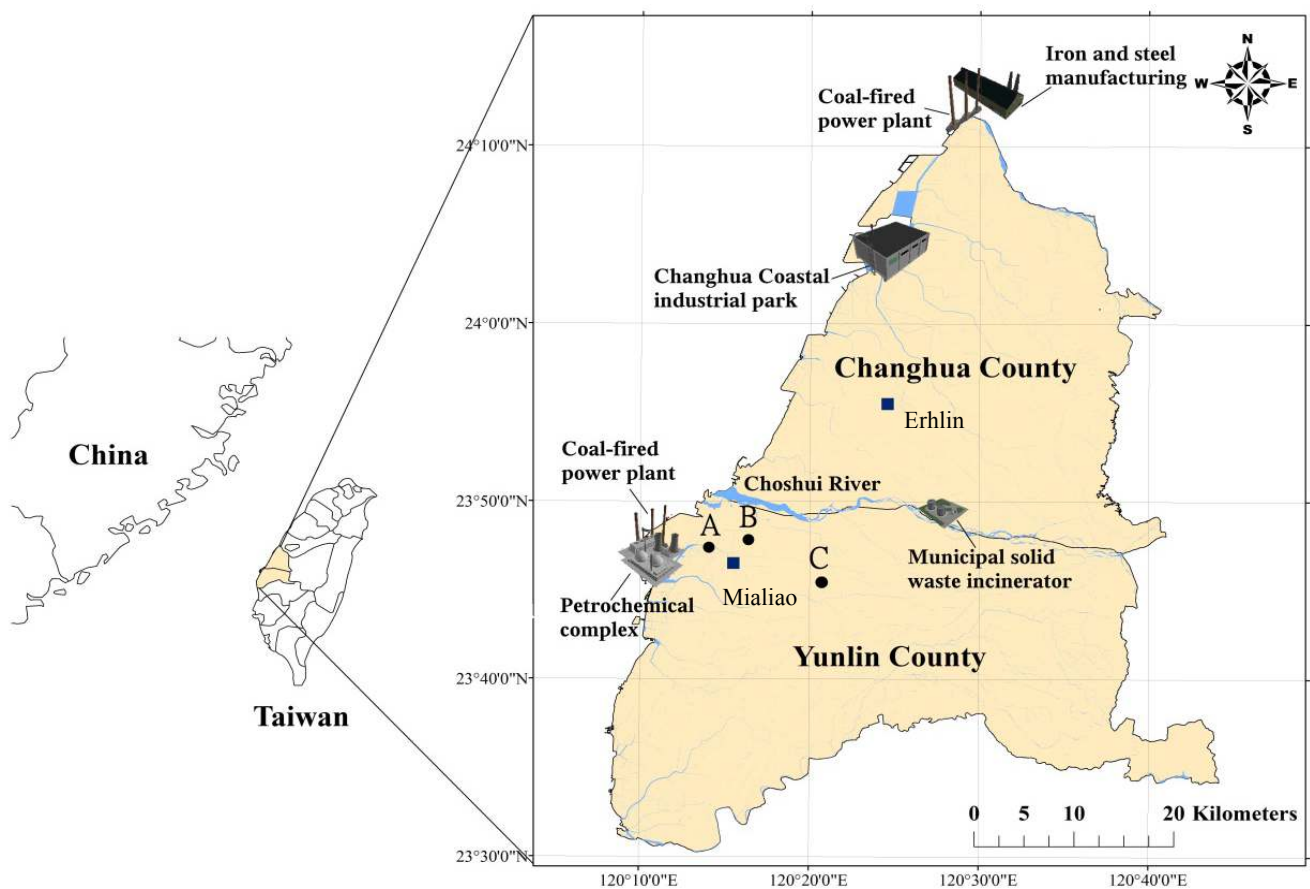
to determine the source profiles and contributions to fine and coarse particles of trace metals based on the knowledge of the variation of elemental concentrations and meteorological parameters.

## METHODS

### Site Description and Sample Collection

Three selected sampling sites (A, B, and C shown in Fig. 1) are located in the suburban in Yunlin County within western coastal central Taiwan. There are several obvious nature and anthropogenic sources of PM and trace metals near the sampling sites, such as ~4 km south of the Choshui River, ~5 to 24 km east of a petrochemical complex (including a coal-fired power plant), within 20 km southwest from a municipal solid waste incinerator and within 60 km south from the Changhua Coastal industrial park (including non-ferrous metal smelting, auto parts manufacturing, glass manufacturing, hardware manufacturing, metal recovery facility, metal production and process, cement production, plastic production, oil refining, and truck and auto painting are main industrial activities), semiconductors manufacturing, coal-fired power plant (Taichung power plant is the largest one in the world), and integrated iron and steel manufacturing.

The period of the air quality deterioration resulted from elevated PM was defined as episode (10th and 11th,



**Fig. 1.** Schematic description of the sampling sites (A: Fong-An, B: Chew-Tow, and C: Lun-Ba) and monitoring stations of Taiwan EPA (■).

December, 2013) and the rest of sampling days was defined as non-episode (9th, 12th and 13th December, 2013). An elevated PM air quality deterioration alert was announced for this region by the Taiwan EPA when local hourly averaged concentrations of PM<sub>10</sub> exceeded 125 µg m<sup>-3</sup>. Here, the typical background level of PM<sub>10</sub> is around 60 µg m<sup>-3</sup> (geometric mean for winter in 2013) obtained from a nearby Mailiao monitoring station of Taiwan EPA (see Fig. 1). Though it was impossible to specifically determine the episode's duration by a daily integrated sampling, we arbitrarily examined the period running from the day (December 10th) that the PM episode was announced to the following day (December 11th), which exhibited an average PM<sub>10</sub> level of > 125 µg m<sup>-3</sup>, as the PM episode period.

The instruments used in this study were installed on the roof (10 m above ground) of three elementary schools (A, B, and C in Fig. 1). All measurements were conducted on five consecutive days from 9th to 13th, December, 2013. Ambient PM<sub>2.5</sub> and PM<sub>10</sub> samples were simultaneously collected by two aerosol samplers (BGI Incorporated, Waltham, MA, U.S.A., Model PQ200) side by side in each of three monitoring sites. Both PM<sub>2.5</sub> and PM<sub>10</sub> samplers with the 47 mm Teflon filters (Whatman PTFE) were operated at the flow rate of 16.7 L min<sup>-1</sup> for 24 hours. One sampler was equipped with a PM<sub>2.5</sub> cut cyclone (Model VSCC), while the other was equipped with a PM<sub>10</sub> cut cyclone. Total volume of air passed through a filter was computed using the flow recorder for each sample. Before and after sampling, the flow rate of each sampler was calibrated by a primary flow meter (MesaLabs Bios Defender 520). Loaded and unloaded filters were dried in a darkened desiccator with the relative humidity of 40% ± 5%.

#### Gravimetric and Chemical Analysis

The PTFE filters were measured gravimetrically for particle mass concentration using an electronic microbalance (Mettler-Toledo MX5) with sensitivity of ± 1 µg after 24 h equilibration at 23 ± 1°C with relative humidity at 40 ± 5%. Each filter was weighted at least three times before and after sampling. Filters were handled only with tweezers coated with Teflon tape to reduce the possibility of contamination. Differences among replicate weights were mostly less than 10 µg for each sample. Net mass was obtained by subtracting pre-weight from post-weight.

Each PTFE filter was cut into two equal halves with ceramic scissors, and then subjected to extraction and digestion. One of half filters was dissolved with acids for elemental measurement. Digestion was performed with an acid mixture (5 mL HNO<sub>3</sub> and 0.3 mL HF) for 90 minutes and 2.8 mL 5% H<sub>3</sub>BO<sub>3</sub> for the other 60 minutes by using an ultrahigh throughput microwave digestion system (Anton-Paar Multiwave 3000). Then the extracts were cooled and diluted to 15 mL with 5% H<sub>3</sub>BO<sub>3</sub>. A blank and two filter blanks were prepared in each run following the same procedures adopted for the samples. All acids used in this study were of ultrapure grade (Merk, Germany). A suit of trace elements, including Magnesium (Mg), Aluminium (Al), Calcium (Ca), Titanium (Ti), Vanadium (V), Chromium (Cr), Manganese (Mn), Iron (Fe), Cobalt (Co), Nickel (Ni), Copper

(Cu), Zinc (Zn), Arsenic (As), Selenium (Se), Strontium (Sr), Molybdenum (Mo), cadmium (Cd), Antimony (Sb), Barium (Ba), and Lead (Pb) were analyzed by inductively coupled plasma mass spectrometry (ICP-MS, Agilent-7700x). The minimum detection limits (MDL) were within the range of 0.001–3.14 µg L<sup>-1</sup> for all selected trace elements. Calibration was achieved using multielement standards prepared from stock standard (Merck) in 2% HNO<sub>3</sub> solution. Quality assurance and control of ICP-MS was guaranteed by the analysis of a certified reference standard, NIST SRM-2786 (urban particulate matter (< 4 µm) with the resulting accuracy of ± 20%. The precision (relative percent difference, RPD) of the instrument was < 10% for the analyzed standards. One field blank with 6–7 field samples was collected for background contamination. The field and filter blanks were analyzed using the same procedure as the samples. There was no significant difference between field and filter blanks. The blank value of each selected elements was subtracted from the analyzed values from the real samples.

#### Data Analysis

In this study, the concentrations and metal compositions on fine particle (PM<sub>2.5</sub>) and coarse particle (PM<sub>2.5-10</sub>) were characterized, where coarse particle was calculated by subtracting PM<sub>2.5</sub> from PM<sub>10</sub>. The measurement data for fine and coarse particles obtained from three sampling sites was integrated to present mean values with variations since there was no between-site variability in concentrations. The meteorological parameters including wind direction, wind speed, temperature, relative humidity (RH) with hourly and 24-hr average were obtained from the EPA monitoring station (Mailiao) of Taiwan which are nearby our sampling sites (Fig. 1).

#### Source Identification Methods

The enrichment factor (EF) analysis which has been widely employed in atmospheric chemistry (Gao *et al.*, 2002; Alleman *et al.*, 2010; Hieu and Lee 2010; Lim *et al.*, 2010; Okuda *et al.*, 2013; Kulshrestha *et al.*, 2014) was used to delineate the source origin (anthropogenic or crustal) of trace metal abundances for atmospheric fine and coarse particles. The EF value is the ratio of concentration ratios of elements in the particle samples normalized to crustal concentrations, where Al is commonly used as crustal source indicator. The EF value can be calculated by using the following equation:

$$EF = (X_i/Al)_{PM} / (X_i/Al)_{Crust} \quad (1)$$

where (X<sub>i</sub>/Al)<sub>PM</sub> is the concentration ratio of a given element X to Al in collected ambient PM and (X<sub>i</sub>/Al)<sub>Crust</sub> is the concentrations ratio of an interested element X to Al in the average crust abundance (Taylor and McLennan, 1995). Considering the uncontaminated background level of soil and the collected PM samples likely relevant to regional and long-range transport particles, the general continental crust (earth's crust) is preferred to use for reducing substantial variation. There is no widely accepted rule for the choice of the reference element; Si, Al and Fe are usually used for

this purpose. In this study Al was used as reference while the composition of the earth's crust was taken from Bowen (1979). This calculation assumes that contribution of anthropogenic Al is insignificant at this sampling region. If the EF value approaches 1, crustal sources are predominant. In general, elements with EF values between 5 and 10 have a significant contribution from non-crustal sources. Elements with high EF values ( $> 10$ ) are essentially completely from anthropogenic origin.

Principal component analysis (PCA) can elucidate variance in a given dataset in terms of minimum number of significant component. Here PCA (SAS version 13.0) was used to perform the source origin based on the selected trace metals for airborne fine and coarse particles. Only the factors with eigenvalues  $> 1$  were selected to identify their associated origins. A factor loading close to one indicated that the element is characteristic of that particular source. Here, a factor loading  $> 0.5$  was adopted to assign source origin to a given principal component. Recently, Positive Matrix Function (PMF) has been widely used in many studies (Viana *et al.*, 2008; Stortini *et al.*, 2009; Tao *et al.*, 2014); however, PMF is not feasible for this study because of lack of required uncertainty factors.

Trajectory analysis has been widely used to interpret air mass transport routes (Yoon *et al.*, 2006). In this study, backward trajectories with an NOAA hybrid single particle lagrangian integrated trajectory (HYSPLIT) model developed by NOAA Air Resources Laboratory (Draxler and Rolph, 2003) were used to determine the transport trajectory of air parcels at the sampling site for 72 hr periods starting at the end of each sampling day. The backward trajectories were starting every 6 hr from 14 m sampling heights, leading to a total of 5 HYSPLIT trajectories within each day.

## RESULTS AND DISCUSSION

### Mass Concentrations of Particles

Table 1 shows the meteorological conditions and air

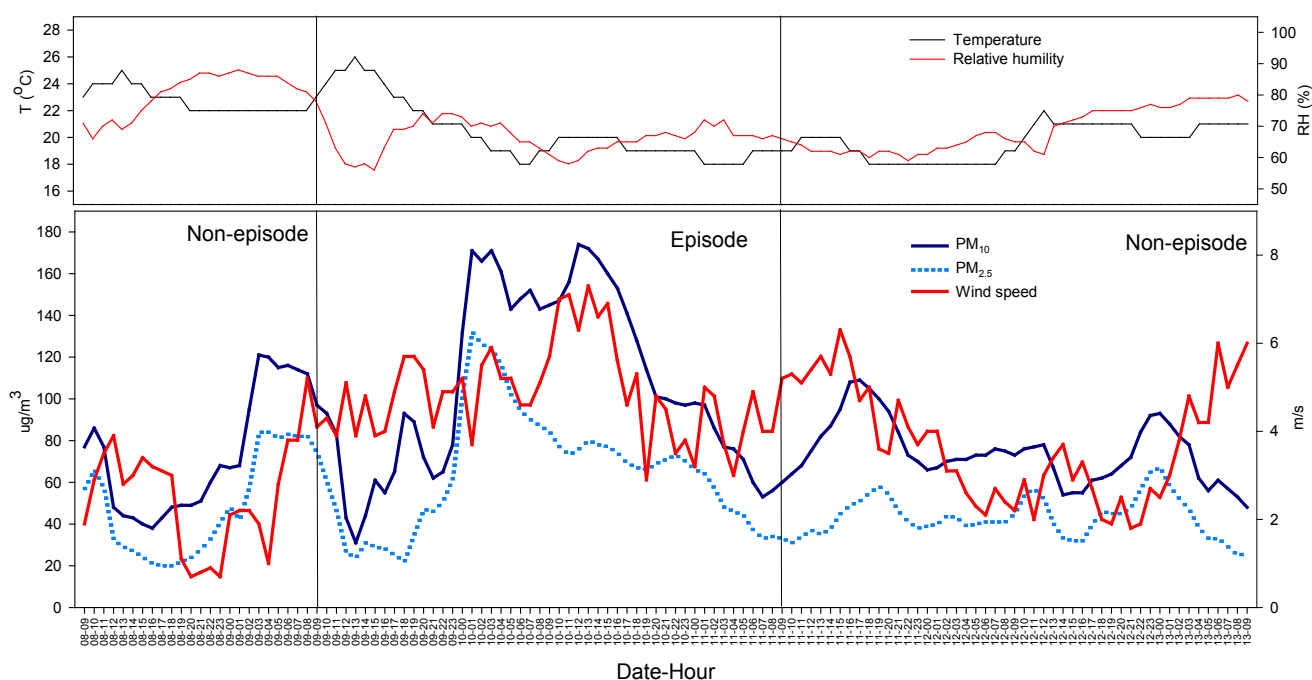
pollutant concentrations (CO, NO<sub>2</sub>, O<sub>3</sub>, SO<sub>2</sub>, PM<sub>10</sub> and PM<sub>2.5</sub>), on average, obtained from a nearby Mailiao monitoring station of Taiwan EPA for episode and non-episode periods. The episode could yield higher mean concentrations of gas contaminants (CO, NO<sub>2</sub>, O<sub>3</sub>, and SO<sub>2</sub>) and particulate matters (PM<sub>10</sub> and PM<sub>2.5</sub>) than non-episode, while no significant difference between periods for temperature, humidity, and wind speed was obtained. The predominant wind direction was northeast for both periods. Since the above averaged values either in air pollutants or meteorological parameters were not adequate to explore detailed information on transportation and dispersion of pollutants, we illustrated the hourly plot of the meteorological conditions (wind speed, temperature and humidity) and PM levels (Fig. 2) and evaluated their associations using a Pearson correlation for both periods. We found the wind speed and PM<sub>10</sub> had similar patterns during the episode, which resulted in a moderate magnitude of correlation ( $r = 0.50$ ). However, poor correlations of the wind speed with PM<sub>10</sub> ( $r = 0.25$ ) for episode and with PM<sub>2.5</sub> ( $r = 0.11$ ) for episode and non-episode ( $r = -0.10$ ) were observed.

The mass concentrations of PM<sub>2.5</sub>, PM<sub>2.5-10</sub>, and PM<sub>10</sub> and ratios of PM<sub>2.5</sub>/PM<sub>10</sub> obtained from PQ200 samplers for the episode and non-episode are also shown in Table 1. The mean concentrations of PM<sub>2.5</sub>, PM<sub>2.5-10</sub> and PM<sub>10</sub> for the episode were higher than those for the non-episode. Mean PM<sub>2.5</sub> concentration for the episode was 1.6 times higher than that for the non-episode while mean PM<sub>2.5-10</sub> and PM<sub>10</sub> concentrations for the episode were about 2.5 and 2 times, respectively, higher than those for the non-episode. The mean ratio of PM<sub>2.5</sub>/PM<sub>10</sub> ( $= 0.43$ ) for the episode was remarkably lower than that for the non-episode ( $= 0.64$ ), indicating that this episode may have generated larger particles and/or promoted the formation of small-particle aggregations. Kuo *et al.* (2013) have indicated that the deterioration of air quality occurred in central Taiwan is usually caused by several types of episode, including long-range transport with dust storms (consisted primarily of mineral dust, but

**Table 1.** The meteorological conditions and air pollutant concentrations (CO, NO<sub>2</sub>, O<sub>3</sub>, SO<sub>2</sub>, PM<sub>10</sub> and PM<sub>2.5</sub>) for episode and non-episode periods.

Station	Parameter	Episode			Non-episode		
		Mean	SD	Range	Mean	SD	Range
Mailiao monitoring station of Taiwan EPA	CO (ppm)	0.63	0.25	0.22–1.15	0.45	0.14	0.20–0.76
	NO <sub>2</sub> (ppm)	17.6	7.12	5.01–32.0	16.5	7.61	5.80–38.0
	O <sub>3</sub> (ppm)	32.1	13.0	5.90–58.0	30.0	13.9	7.20–62.0
	SO <sub>2</sub> (ppm)	7.58	4.48	1.30–20.0	5.96	4.12	1.10–18.0
	PM <sub>10</sub> (μg m <sup>-3</sup> )	107	42.6	31.0–174	74.3	22.2	38.0–121
	PM <sub>2.5</sub> (μg m <sup>-3</sup> )	64.9	28.1	22.0–132	45.1	16.8	20.0–84.0
	Temperature (°C)	21.1	2.09	18.3–26.1	19.0	1.30	18.1–27.4
	Humidity (%)	69.6	9.06	56.1–88.0	70.3	6.73	59.6–81.2
	Wind direction	Northeast	-	-	Northeast	-	-
	Wind speed (m s <sup>-1</sup> )	4.61	1.40	1.00–7.30	4.12	1.36	1.80–6.74
In-situ PQ200 sampler	PM <sub>2.5</sub> (μg m <sup>-3</sup> )	53.8	10.8	40.6–69.0	34.8	10.1	24.6–56.1
	PM <sub>2.5-10</sub> (μg m <sup>-3</sup> )	71.2	29.7	46.2–111	28.5	21.6	25.1–74.2
	PM <sub>10</sub> (μg m <sup>-3</sup> )	121	30.1	90.5–167	63.8	16.4	45.5–98.8
	PM <sub>2.5</sub> /PM <sub>10</sub> ratio	0.43	0.11	0.30–0.54	0.64	0.21	0.42–0.95

SD: standard deviation.



**Fig. 2.** Hourly temperature and humidity profiles and plots of PM level ( $PM_{2.5}$  and  $PM_{10}$ ) and wind speed obtained from a Mailiao monitoring station of Taiwan EPA.

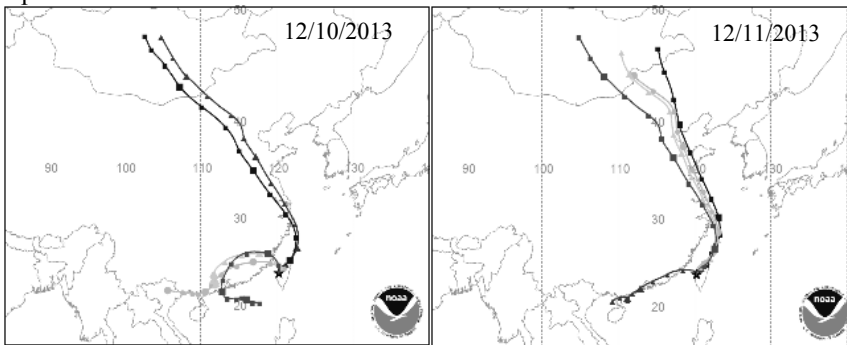
not necessarily air pollutant such as  $SO_2$ ,  $NO_x$  and CO), long-range transport with frontal pollution (consisted of significant amounts of anthropogenic air pollutants from the Asian continent, but not necessarily mineral dust) (Lin *et al.*, 2004), river dust, stagnant weather, and a mixture of episode. The river dust episode with  $PM_{2.5}/PM_{10}$  ratio below 0.5 was reported (Kuo *et al.*, 2010), where coarse particles in the river sand are easily re-suspended by wind during drought seasons (i.e., winter) in central Taiwan. This observed PM episode was also unlikely attributable to a typical local pollution with stagnant weather (wind speed below  $2 \text{ m s}^{-1}$ ) in central Taiwan (Kuo *et al.*, 2013), because the high level of wind speed ( $> 4 \text{ m s}^{-1}$ ) and low ratio of  $PM_{2.5}/PM_{10}$  ( $< 0.5$ ) were observed (see Table 1). Furthermore, we observed air pollutants ( $CO$ ,  $O_3$ ,  $NO_2$ , and  $SO_2$ ) obtained from regional Taiwan EPA station (Erhlin, located the upwind of Choshui River, see Fig. 1) increased to 3–34%, on average, during the episode period as comparing with the non-episode period (not shown). However, only a slight increase in  $PM_{10}$  during the episode period (from  $45.0$  to  $52.5 \mu\text{g m}^{-3}$ , on average) was found at the regional Taiwan EPA station (Erhlin), while the extremely high level of  $PM_{10}$  during the episode period (from  $74.3$  to  $107 \mu\text{g m}^{-3}$ , on average) was measured at the local Taiwan EPA station (Mailiao). The similar meteorological conditions were measured from both Taiwan EPA stations during the sampling period. The above result indicates that Mailiao station had higher  $PM_{10}$  background levels (resulting from local emissions) than Erhlin station within the episode period. Through the backward trajectory analysis using the HYSPLIT model (Fig. 3), we could initially infer this episode was likely caused by the long-range transport with frontal pollution. The contribution of local river/road dusts

to our collected samples, in part, might be taken into account due to dramatically increasing  $PM_{10}$  concentrations. It should be noted that the northeast winter monsoon with particle pollutants passes over the industrial regions of north continental China and Taiwan, which may allow air passels to bring imported and regional air pollutants through the atmosphere into the study area if vertical mixing is favorable to such entrainment (Lin *et al.*, 2004).

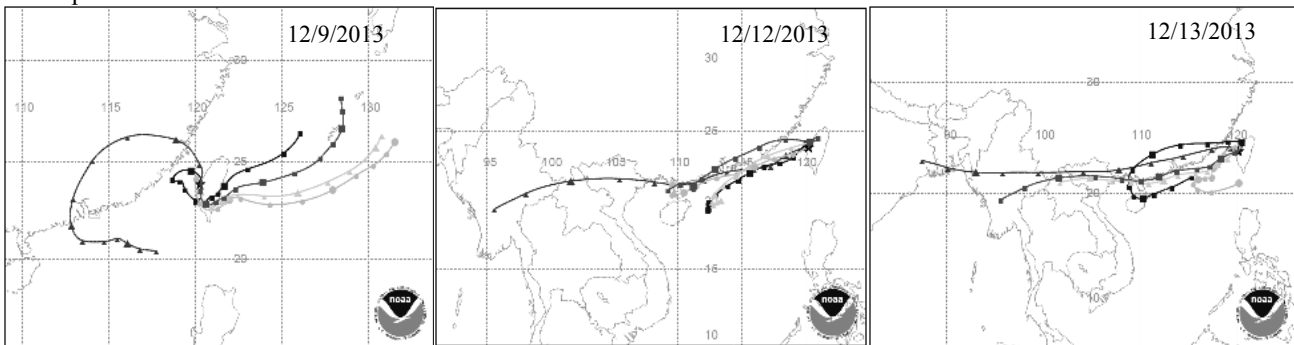
#### Trace Metal Compositions

Table 2 shows the mean concentrations of trace metals in fine and coarse particles for the episode and non-episode periods. For episode, in addition to Al, Ca, Fe, Mg, and Zn ( $100$  to  $1200 \text{ ng m}^{-3}$ ), Pb, Cr, Ti, Mn, and Cu ( $10$  to  $100 \text{ ng m}^{-3}$ ) were also major metals in the fine particle, followed by Ni, Ba, V, As, Sb, Sr, and Cd. The rest of elements with concentrations less than  $1 \text{ ng m}^{-3}$  were Mo, Se, and Co. In the coarse particle, Al, Fe, Ca, Mg, Ti, and Zn with a high concentration ( $> 100 \text{ ng m}^{-3}$ ) were observed, while Mn, Ba, Pb, Cr, Cu, Sr, V, Ni, As, Co, and Se had moderate abundance in concentrations ( $1$ – $50 \text{ ng m}^{-3}$ ). Mo, Sb and Cd were at even lower concentrations ( $< 1 \text{ ng m}^{-3}$ ). For non-episode, Al was also the most abundant element in the fine fraction, followed by Ca, Mg, Fe, and Zn ( $100$ – $1000 \text{ ng m}^{-3}$ ). The moderate concentrations ( $10$ – $100 \text{ ng m}^{-3}$ ) for Pb, Cr, Ti, Mn, and Cu were obtained, followed by V, Ba, Ni, As, Sb, Sr, Se, and Cd with concentrations ranged from  $1$  to  $10 \text{ ng m}^{-3}$ . The trace elements with concentrations below  $1 \text{ ng m}^{-3}$  were Mo and Co. In the coarse particle, the most abundant elements were Al, Ca, Fe, and Mg with concentrations ranged from  $100$  to  $1000 \text{ ng m}^{-3}$ . The moderate concentrations ( $10$ – $100 \text{ ng m}^{-3}$ ) for Ti, Zn, Cr, Ba, Mn, and Pb were recorded, followed by Cu, Sr, Ni, V, As, Sr, and Sb ( $1$ – $10 \text{ ng m}^{-3}$ ),

## Episode



## Non-episode



**Fig. 3.** Backward trajectories of the study area for 5 sampling days.

**Table 2.** The concentration ( $\text{ng m}^{-3}$ ) of selected trace metals in ambient fine ( $\text{PM}_{2.5}$ ) and coarse ( $\text{PM}_{2.5-10}$ ) particles for episode and non-episode periods.

Metals	Episode				Non-episode			
	$\text{PM}_{2.5}$ (n = 6)		$\text{PM}_{2.5-10}$ (n = 6)		$\text{PM}_{2.5}$ (n = 9)		$\text{PM}_{2.5-10}$ (n = 9)	
	mean	SD	mean	SD	mean	SD	mean	SD
Mg	344	28.9	726	531	334	44.3	449	162
Al	1136	158	3129	2870	999	38.6	976	764
Ca	739	97.1	1035	521	657	252	691	297
Ti	32.5	8.22	199	168	20.5	3.90	64.2	35.2
V	6.87	1.11	10.1	5.41	6.20	2.12	3.65	1.96
Cr	36.8	20.4	16.9	8.68	26.8	12.4	27.2	16.2
Mn	28.6	1.82	46.8	22.4	16.7	5.02	14.9	8.04
Fe	427	72.5	2155	1723	198	38.0	666	349
Co	0.435	0.061	1.13	0.775	0.251	0.047	0.524	0.29
Ni	9.14	4.94	5.345	3.34	5.65	1.90	4.88	2.97
Cu	18.1	1.09	15.1	8.79	12.5	3.47	8.761	3.38
Zn	202	19.2	107	31.9	133	55.9	68.2	47.8
As	6.67	1.46	2.10	1.50	3.45	1.04	1.12	0.607
Se	0.776	0.552	1.08	0.054	1.18	0.927	1.02	0.749
Sr	3.65	0.225	11.5	7.27	2.45	0.839	5.96	1.94
Mo	0.979	0.196	0.957	0.194	0.645	0.397	0.779	0.489
Cd	1.90	0.512	0.604	0.225	1.04	0.330	0.317	0.217
Sb	4.91	0.337	1.41	0.696	3.29	0.537	1.04	0.402
Ba	8.25	0.612	33.9	24.3	5.67	1.20	18.2	11.1
Pb	78.7	15.8	23.8	9.87	33.9	13.8	13.2	7.62

n: number of measurements; SD: standard deviation.

while Mo, Co, and Cd with the lowest concentrations ( $< 1 \text{ ng m}^{-3}$ ) were observed.

When the PM episode occurred, we found mean levels

of selected trace metals (exception for Se) in the fine particle were increased to 3–132%, where Pb, As and Fe levels were increased to 132%, 116% and 93%, respectively,

in comparison with non-episode. Besides, mean levels of coarse-size trace metals (exception for Cr) were increased to 6–223% during the episode period, where Fe, Al, Mn, Ti, and Co levels were significantly increased to 223%, 220%, 214%, 210%, and 115%, respectively. Here the ratios of fine/coarse metals for Cr, Ni, Cu, Zn, As, Mo, Cd, Sb, and Pb for the episode were greater than 1. In addition to above metals, V, Mn, and Se obtained from the non-episode also had a ratio of fine/coarse metals greater than 1. Gemenetzi *et al.* (2006) have indicated that high fine/coarse (or  $PM_{2.5}/PM_{10}$ ) element ratios are associated with combustion-related elements, such as Pb, Zn, Cd, and V. A comparison of the average values for As, Cd, Ni, Pb with limit values fixed by European directive shows that the value of fine-size As ( $6.67 \text{ ng m}^{-3}$ ) obtained from the episode is above the limit level ( $6 \text{ ng m}^{-3}$ ) of European directive (2004/107/EC), while the levels for fine-size Cd, Ni, and Pb do not exceed.

Kuo *et al.* (2010) had conducted a study with particle measurements for the river dust episode in Yunlin County where the crustal metals (Fe + Al + Mn + Ca) in fine and coarse particles during the non-episode period, on average, were  $1.1$  and  $2.2 \text{ } \mu\text{g m}^{-3}$ , respectively. Those concentrations are comparable to our results of fine- ( $1.3 \text{ } \mu\text{g m}^{-3}$ ) and coarse-size ( $2.0 \text{ } \mu\text{g m}^{-3}$ ) crustal metals obtained from the non-episode period. However, our results of crustal metals (fine = 2.33, coarse =  $6.39 \text{ } \mu\text{g m}^{-3}$ , on average) from the long-range transport episode were higher than those (fine = 1.92, coarse =  $5.27 \text{ } \mu\text{g m}^{-3}$ , on average) from the river dust

episode. These two types of episode could yield different ratios of crustal elements (Fe or Al)/reference elements (Cd or Mo) as indexes used to distinguish particle characteristics, indicating that their contribution sources of particulate metals are differed. Moreover, Hsu *et al.* (2004) reported that a moderate Cd/Pb (0.024) and Zn/Pb (1.98) ratio for a northeast monsoon period was obtained from northern Taiwan, while a low Cd/Pb (0.017) and Zn/Pb (1.82) ratio for the Asian dust period was observed. Those ratios for a northeast monsoon period are similar to our observed episode's results (Cd/Pb = 0.024 and Zn/Pb = 2.63) but lower than findings (Cd/Pb = 0.033 and Zn/Pb = 4.05) from the non-episode period. According to aforementioned comparisons of local/regional air pollutants, the backward trajectory, and metal fingerprints, we could further confirm that the observed episode was attributed to the long-range transport with frontal pollution (northeast winter monsoon), which subsequently increase higher PM levels, in spite of similar meteorological conditions between two periods.

#### Enrichment Factor

Fig. 4 shows the results (mean and standard deviation) of enrichment factor analysis for all analyzed elements in the fine and coarse size ranges obtained from the episode and non-episode. The mean EF values of Ti, Fe, and Sr in the fine particles approached unity for both periods, indicating that these elements originated mainly from crustal sources. Ba, Mn, Ca, Co, Mg, V, and Ni with EFs between 1 and 10 were from crustal and non-crustal sources. Cd and Sb were

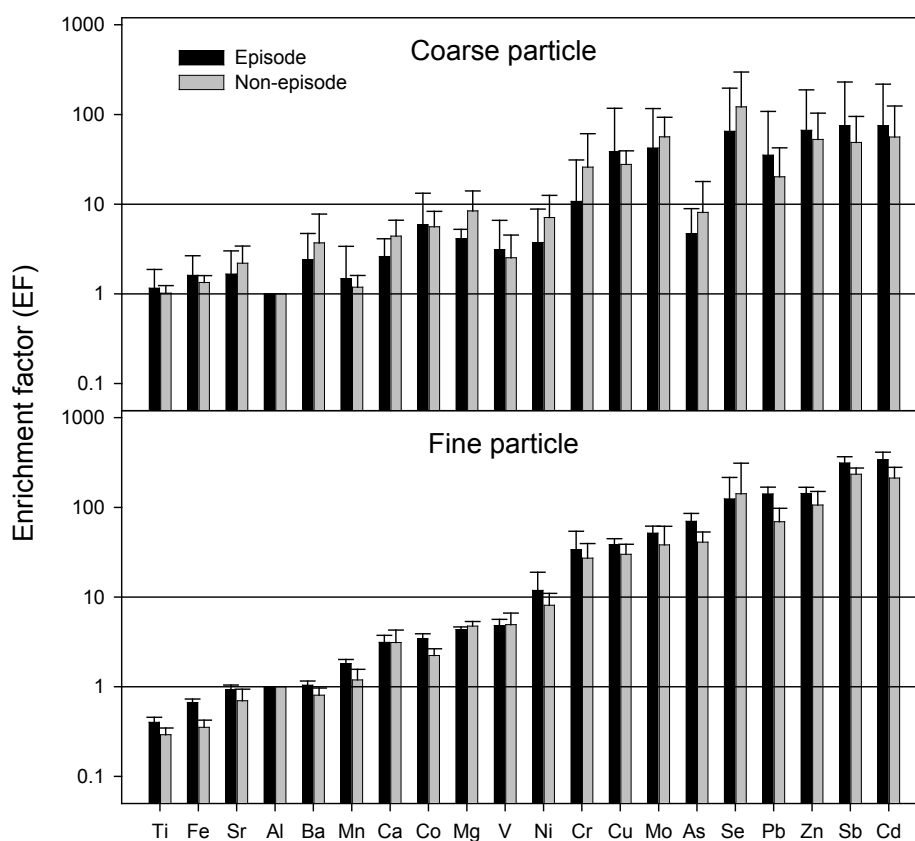


Fig. 4. The EF values for trace metals in fine and coarse particles obtained from episode and non-episode periods.

the most enriched elements, followed by Zn, Pb, Se, As, Mo, Cu, and Cr with mean EFs > 10, indicating that these trace metals originated mainly from anthropogenic sources.

For coarse particles during the periods of the episode and non-episode, the mean EF for Ti was less than 1, indicating this metal originated from soil; whereas the EFs for Cr, Cu, Mo, As, Se, Zn, Pb, Sb and Cd were much higher than 10, indicating an anthropogenic origin of those metals. The moderate EFs of the remaining metals (Fe, Sr, Ba, Mn, Ca, Co, Mg, V and Ni) indicated crustal and non-crustal sources. The lower EFs with increasing particle size for Sb, Se, As, Mo, Cr, Cu, and V were found for both periods in this study, in addition to Pb, Zn, Ni and Cd reported by previous studies (Baeyens and Dedeurwaerder, 1991; Chan *et al.*, 1997; Manoli *et al.*, 2002; Hieu and Lee 2010; Hu *et al.*, 2013). It is worth noting that the top four anthropogenic metals (Cd, Sb, Pb and Zn) with high EFs in the ambient fine particle were also observed in Taipei city, northern Taiwan (Hsu *et al.*, 2002; Gugamsetty *et al.*, 2012).

The total mean concentrations of trace metals (such as Cd, Sb, Zn, Pb, Se, As, Mo, Cu, Cr, and Ni) with non-crustal sources in fine and coarse particles for the episode were ~1.5 times higher than those for the non-episode. Furthermore, the total mean concentration of crustal metals (Al, Fe, Mg, and Ca presenting higher levels) in the fine particle for the episode was about 1.2 times higher than those for the non-episode while their levels in the coarse particle were about 2.5 times higher than those for the non-episode. The similar patterns with those crustal metals in fine and coarse particles resulted from the river-dust episode in central Taiwan have been reported (Kuo *et al.*, 2010). Our results show that the episode caused by the long-range transport with frontal pollution could not only increase the magnitude of crustal elements, but also bring the abundant metals from anthropogenic sources.

#### Source Apportionment Using PCA

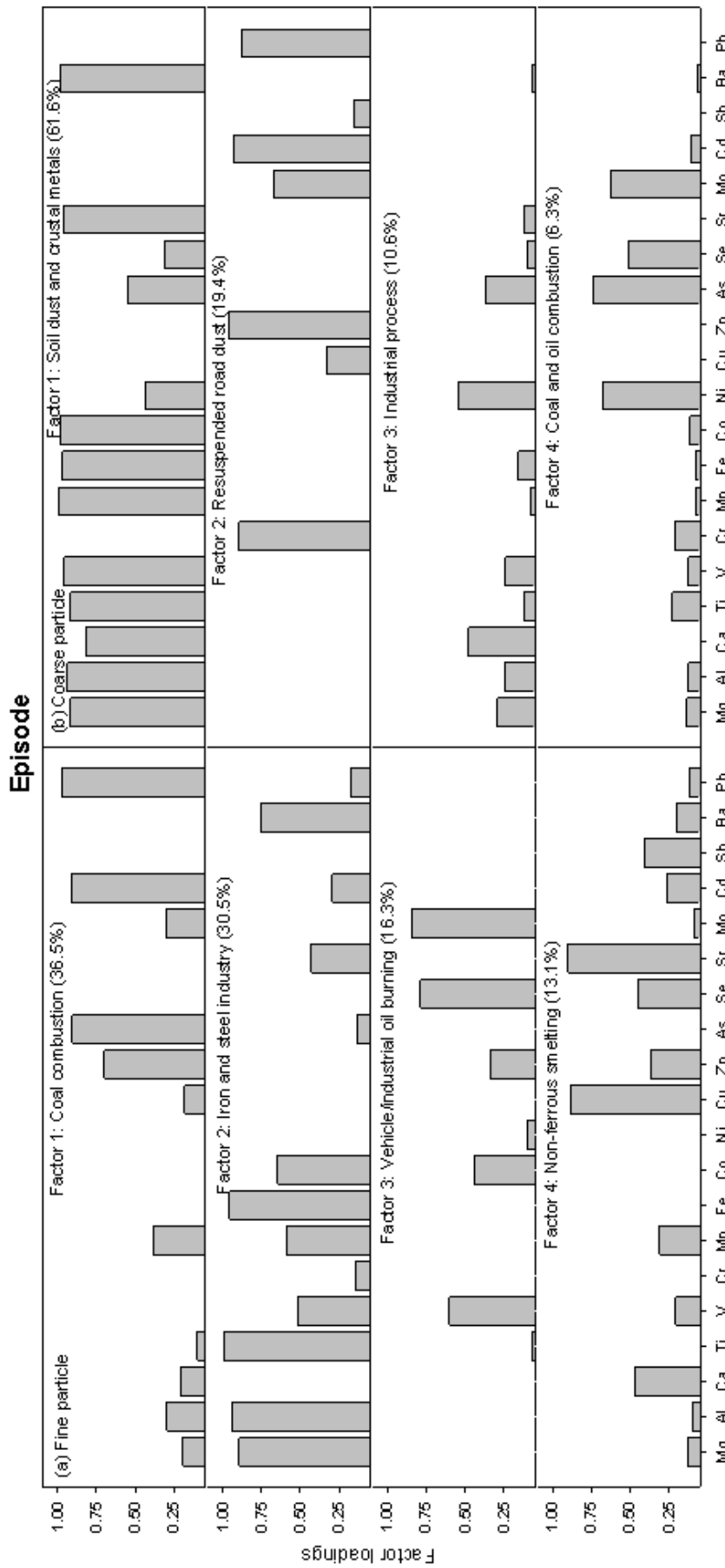
The rotated factor loading of trace metals in the fine and coarse particles for episode and non-period are presented in Figs. 5 and 6, respectively. As shown in Fig. 5, four factors accounted for 96.5% of the accumulative variance in fine particles for the episode. Factor 1, explaining 36.5% of the total variance, is strongly correlated with Zn, As, Cd, and Pb. This factor is likely to refer to coal combustion, where As is usually identified as tracer for coal burning (Harrison *et al.*, 1996). However, As is sometimes attributable to industrial process, such as the manufacturing of semiconductors, electronics and electrical peripheral, which intensively located in northern and central Taiwan (Chen *et al.*, 2013). Factor 2, accounting for 30.5% of the total variance, comprises Mg, Al, Ti, Fe and Ba with high loadings and Co, Mn and V with moderate loadings (0.5–0.7). This factor refers to iron and steel industry (Querol *et al.*, 2007; Tsai *et al.*, 2007). Factor 3 explaining 16.3% of the total variance is correlated with high loadings on Mo and Se and moderate loadings on V, Zn and Co. This factor is associated with vehicular emission and oil burning (Fung and Wong 1995; Stortini *et al.*, 2009). Factor 4 explaining 13.1% of the total variance showed high loadings on Cu, and Sr, which is

associated with non-ferrous metallurgy, such as Cu smelting source (Querol *et al.* 2007; Viana *et al.* 2008). There are several non-ferrous smelters located in central Taiwan. In coarse particles, four factors account for 97.9% of the overall variance. Factor 1 (explaining 61.6% of the total variance) includes Mg, Al, Ca, Ti, V, Mn, Fe, Co, Sr, and Ba which refer to soil dust and crustal elements (Manoli *et al.*, 2002; Minguillón *et al.*, 2012). Factor 2 (explaining 19.4% of the total variance), containing Zn, Cd, Pb, and Mo, refers to traffic-related re-suspended road dust (Weckwerth, 2001; Kuo *et al.*, 2010). Factor 3 explaining 10.6% of the total variance has moderate loadings on Ca, Mg, Al, Ni, and As, which is considered as industrial process. Factor 4 (explaining 6.3% of the total variance) includes Se, Mo, Ni, and As which refer to coal and oil combustions. In this case, As and Se have been reported as tracers of coal combustion (Swietlicki *et al.*, 1996; Manoli *et al.*, 2002), while Ni and Mo are considered as oil combustion (Swietlicki *et al.*, 1996; Manoli *et al.*, 2002; Singh *et al.*, 2002; Al-Momani, 2003).

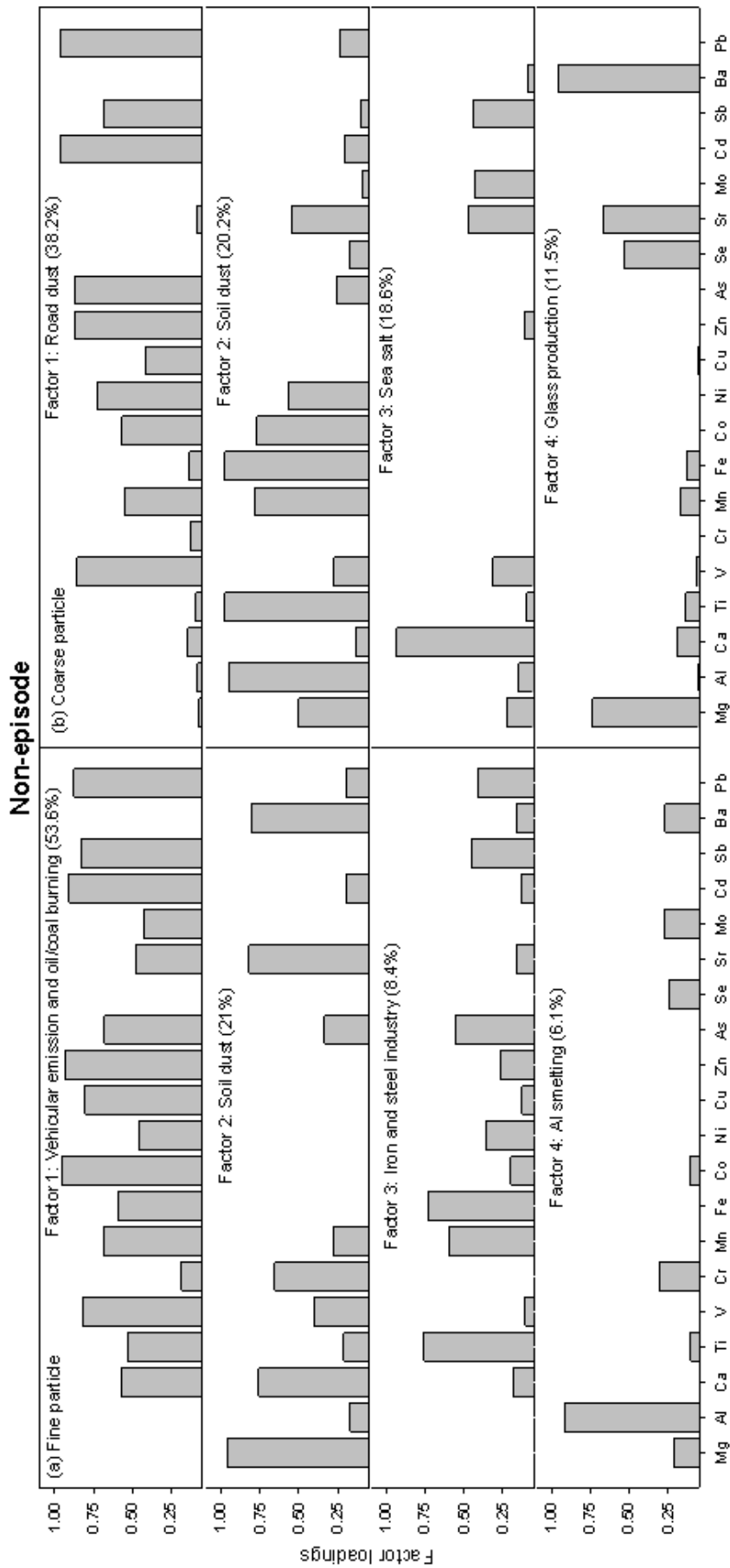
For the non-episode (Fig. 6), four factors account for 89.4% of the accumulative variance in fine particles. Factor 1 includes V, Co, Cu, Zn, Cd, Sb, and Pb with high loadings and Ca, Ti, Mn, Fe, and As with moderate loadings, explaining 53.6% of the total variance. This factor is likely to associate with mixed sources of vehicular emissions and oil/coal burning (Lin *et al.*, 2005; Wang *et al.*, 2003; Lee *et al.*, 2011). Factor 2, explaining 21% of the total variance, has high loadings on Mg, Ca, Sr, and Ba and a moderate loading on Cr. This factor is associated with soil dust and crustal metals (Swietlicki *et al.*, 1996; Marcazzan *et al.*, 2001; Singh *et al.*, 2002; Al-Momani, 2003). Factor 3 accounting for 8.4% of the total variance is related to Ti, Fe, Mn, Ni, As, Sb, and Pb. This factor refers to iron and steel industry (Mansha *et al.*, 2012; Dall'Osto *et al.*, 2008). Factor 4 includes Al, Cr, Se, and Mo, explaining 6.1% of the total variance. This factor is probably associated with Al smelting (Querol *et al.*, 2007). In coarse particles, four factors also account for 88.5% of the overall variance. Factor 1 (38.2%) comprises V, Mn, Co, Ni, Cu, Zn, As, Cd, Sb, and Pb referring to traffic-related re-suspended road dust. Factor 2 (20.2%) includes Mg, Al, Ti, Mn, Fe, Co, Ni, and Sr, which is considered as soil dust and crustal metals. Factor 3 (18.6%) with a high loading on Ca and moderate loadings on Sr, Mo and Sb refers to sea salt. Factor 4 (11.5%) with high loadings on Mg, Se, Sr and Ba is associated with Glass production.

While we successfully identified the anthropogenic and natural sources (such as industrial emissions, traffic, re-suspended road dusts and crustal metals) of trace metals in fine and coarse particles for the episode or non-episode in the study site using a PCA, we were not able to strictly distinguish the contribution of local or imported PM metals. That is because that the imported air parcels containing PM metals with long-range transport and winter monsoon pass over the industrial sources (i.e., coal-fired power plant) of continental China or Taiwan and may pick up its emitted air pollutants to our study area. As a result, coal combustion (as an example) is one of identified emission sources using a PCA but belonging to local, regional and/or imported sources is unclear. Thus, it might be a challenge in PM source





**Fig. 5.** Factor analysis patterns after normalized varimax rotation of trace metals and four identified sources for fine (a) and coarse (b) particles for the episode.



**Fig. 6.** Factor analysis patterns after normalized varimax rotation of trace metals and four identified sources for fine (a) and coarse (b) particles for the non-episode.

identification for further control strategies if the study area of interest is influenced by multiple sources or episode.

## CONCLUSIONS

The distribution of particle mass and trace metal concentrations in the fine and coarse particles for the periods of episode and non-episode was investigated at the suburban area, in Yunlin County of central Taiwan. Although we arbitrarily separated the episode and non-episode periods by a daily integrated sampling, the results on backward trajectories, PM mass levels and associated metal concentrations between periods were significant and reasonable. The mean concentrations of PM<sub>2.5</sub>, PM<sub>2.5–10</sub>, and PM<sub>10</sub> obtained from the episode period were 53.8, 71.2 and 121  $\mu\text{g m}^{-3}$ , respectively while the relatively lower concentrations of PM<sub>2.5</sub> (34.8  $\mu\text{g m}^{-3}$ ), PM<sub>2.5–10</sub> (28.5  $\mu\text{g m}^{-3}$ ), and PM<sub>10</sub> (61.9  $\mu\text{g m}^{-3}$ ) were observed for non-episode. Al, Fe, and Ca were predominant in fine and coarse particles for both periods, accounting for 75–85% of all selected elements. When the PM episode occurred, we found that mean concentrations of fine-size Pb and As were increased to 132% and 116%, respectively, while average concentrations of coarse-size Fe, Al, Mn, Ti, and Co were increased to 223%, 220%, 214%, 210%, and 115%, respectively, in comparison with non-episode. Our results also showed the EFs were decreased with increasing particle size for Sb, Se, As, Mo, Cr, Cu, V, Pb, Zn, Ni and Cd in both periods. This PM episode could generate higher EFs for all identified anthropogenic elements in fine particles. As a result, the mean value of fine-size As (6.67  $\text{ng m}^{-3}$ ) obtained from the episode period was higher than the proposed European Union standard (6  $\text{ng m}^{-3}$ ). Through principal component analysis, the anthropogenic and natural sources of fine- and coarse-size metallic elements for the episode and non-episode were identified, such as industrial emissions, traffic and soil dust/crustal metals. We suggest that the emission from coal combustion has to be well controlled for reducing health risks of residents living in our study area because of its high contribution (magnitude and associated toxic metals, such as As) to the atmospheric fine particle. This study provides essential information for conducting health and environmental impact assessments and initiating further control strategies.

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