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Ambient air particulate concentrations and metallic elements principal component analysis at Taichung Harbor (TH) and WuChi Traffic (WT) near Taiwan Strait during 2004–2005

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Abstract

The purpose of this study is to characterize metallic elements associated with atmospheric particulate matter of total suspended particulate (TSP), fine particle (particle matter with aerodynamical diameter <2.5 μm, $PM_{2.5}$), coarse particle (particle matter with aerodynamical diameter 2.5–10 μm, $PM_{2.5–10}$) at the Taichung Harbor (TH) and WuChi Traffic (WT) sampling site of central Taiwan during March 2004 to February 2005. The result indicated the average total suspended particulate concentration in 1 year was 157.31 and 112.58 μg m^{-3} at TH and WT sampling site, respectively. Fine particle ($PM_{2.5}$) size was the dominant species at TH and WT sampling site. In TH sampling site, higher correlation coefficient was observed on total suspended particulates of metallic elements Fe and Zn. And in WT sampling site, higher correlation coefficients displayed on total suspended particulates of metallic elements Fe and Zn, Fe and Mn. Ambient airborne particle principal component analysis of metallic metals was used to identify the possible pollutant sources in this study. At the TH sampling site, 50.81% of the total variance of the data was observed in factor 1. Higher loading of Fe (0.86), Zn (0.79), Pb (0.76), and Mn (0.68) were contributed by traffic emission and the soil source. At the WT sampling site, factor 1 explained 53.74% of the total variance of the data and had high loading for Zn (0.86) and Cu (0.85), which were identified as industrial/traffic emission sources.

Keywords: $PM_{2.5}$; $PM_{2.5–10}$; Harbor; Metallic elements

1. Introduction

The coastal marine atmosphere adjacent to large urban and industrial centers can be strongly impacted by pollution emissions, resulting in high loading of pollutants in the ambient air [1–7]. Suspended particle pollution has become a serious problem in Taiwan. Emissions of anthropogenic air pollutants in Asia have been increasing drastically in past decade [8]. According to previous study particulate matter has a potential adverse health effect, making it necessary to control or regulate these pollutants. The sources, characteristics, and potential health effects of the larger (coarse) particles (diameter in 2.5–10 μm) and smaller (fine) particles (diameter smaller than 2.5 μm) are different. Fine particulate not only readily penetrate into the lungs but also are likely to increase respiratory and mutagenic diseases [9]. The health-related finds of these studies were associated with either the total mass concentration of suspended particulate (TSP) or the mass concentration of particles with aerodynamic diameters smaller than 10 μm (PM10) or smaller than 2.5 μm (PM2.5). The concentration, composition, and particle size of suspended particulate matter at a given site are determined by such factors as meteorological properties of the atmosphere, topographical influences, emission sources, and by particle parameters such as density, shape, and hygroscopicity [10].

Atmospheric dry deposition contaminants are known to be an important removal mechanism of trace metals. Despite its important, understanding of dry deposition to natural surface is far from complete. This technique can be used to directly assess deposited material and allows comparisons measured and modeled data [11,12].

Epidemiological studies conducted in several countries show consistent associations of exposure to ambient particulates with adverse health effects including increased mortality, hospitalization for respiratory or cardiovascular disease, and respiratory

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symptoms and decreased lung function. Based on epidemiological time series studies, dose-response functions were identified between an increase in PM and adverse health effects [13].

Ambient air particulate concentrations were performed at Taichung Harbor and WuChi Traffic near Taiwan Strait in central Taiwan during March 2004 to February 2005. The purposes of this study were investigated, characterized ambient air particulate concentrations and those metallic elements associated with them at Taichung Harbor and WuChi Traffic sampling sites.

2. Materials and method

2.1. Sampling sites

Taichung Harbor was an artificial harbor and has the maximum amounts of 83 ports, which is located on the west coast of central Taiwan. The boundaries of the Port of Taichung are thus: The northern boundary is the south beach of Ta-Chia stream, the southern boundary is the north beach of Ta-Tu stream, eastern boundary is Ling Kung Road, and western boundary is the Taiwan Strait. The length of the port is 12.5 km, width is 2.5–4.5 km, the area is 3,760 ha, among them the land area is 2,800 ha, and the water area is 960 ha; this includes three parts to execute the construction of business port, industrial port and fishing port. Another sampling site is located at Chung-Chi Road in front of WuChi Elementary School in central Taiwan. Chung-Chi Road is a main traffic road that connected Taichung Harbor to other cities.

The sampling height of this sampling site is about 10 m. The sampling sites were at HUNG SUN building, which was near the chemical port and at WuChi Elementary building. They were located on the east side of Taiwan Strait. The purpose of this study was to investigate the ambient air, fine, coarse particulate concentrations, dry deposition and those metallic elements which attached with them at Taiwan harbor near Taiwan Strait (Fig. 1).

2.2. Sampling program

2.2.1. Sample collection

2.2.1.1. PS-1 sampler. The PS-1 (GPS1 PUF Sampler, General Metal Work) sampler is a complete air sampling system designed to simultaneously collect suspended airborne particles at flow rate up to 280 l/min and flow rate was adjusted to 200 l/min in this study. The quartz filter (diameter 10.2 cm) is used to filter the suspended particles in the study. The filters were first conditioned for 24 h under an electric chamber at humidity 50 ± 5% and temperature 20 ± 5 °C prior to both on and off weighing. Filters were placed in sealed CD box during transport and storage process.

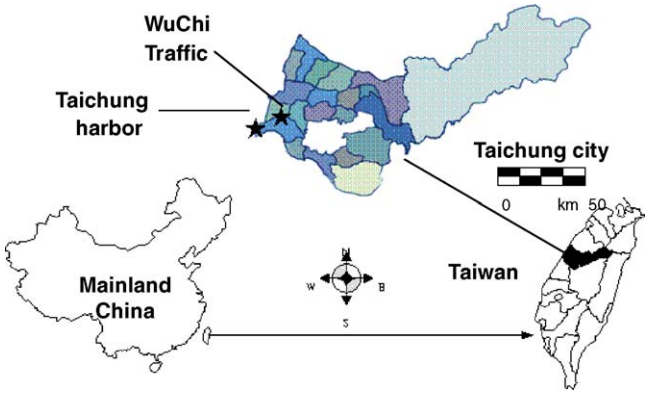

Fig. 1. The location of sampling sites for Taichung Harbor and WuChi Traffic near Taiwan Strait in central Taiwan.

2.2.1.2. Universal air sampler. The Model 310 Universal Air Sampler™ (UAS™) is a general-purpose air sampler for atmospheric aerosol sampling and mass concentration determinations, as well as organic or inorganic sampling. The sampler has a designed inlet sampling flow rate of 300 l/min. It is provided with an omni-directional inlet, a PM_{10} (10 μm cut) virtual impact classifier, either a $PM_{2.5}$ or PM_{10} virtual impact classifier, or a fine particle filter. This allows the sampler to be operated as a high volume dichotomous sampler for size fractionation of airborne particle in the 0–2.5 and 2.5–10 μm aerodynamic size ranges. Air is aspired at 300 l/min from the ambient atmosphere. Particles greater than 10 μm aerodynamic equivalent diameters are removed from the air stream by the PM_{10} classifier and discarded. Particles lesser than 10 μm in diameter are flowed to the $PM_{2.5}$ classifier located downstream. Particles in the 2.5–10 μm range (coarse fraction) are collected on a 62 mm × 165 mm filter and those smaller than 2.5 μm (fine fraction) are collected on a 200 mm × 250 mm final filter.

2.2.2. Meteorological analysis

Meteorological analysis was made by WatchDog weather station Model 525 (Spectrum Technologies Inc., USA). The weather station can provide data of wind speed, wind direction, temperature and humidity during sampling period.

2.2.3. Chemical analysis

After final weighing, all quartz filters by PS-1 and UAS sampler were both cut into one-eighth. Then they were put into 200 ml bottles for each sampling group. Than 10 ml ultrapure HNO_3 was added to digest these particulates at 200–250 °C for 2 h. After above procedure this solution was added with distilled-deionized water to 30 ml. A Hitachi Z-5000 series polarized Zeeman flame atomic absorption spectrophotometer was used to measure the trace metal concentration after digestion process.

2.2.4. Statistical analysis

In this study, a statistical analysis was applied to the particulate data (TSP, $PM_{2.5–10}$ and PM_{10}) and the meteorological data (temperature, relatively humidity, wind speed, and wind direction) collected during the study period. A non-parametric (Spearman) correlation analysis is taken to investigate the correlation coefficients of metallic metal and the particulate correlate with the meteorology data. In addition, the regression analysis is used in this paper. The particulate matters (TSP, $PM_{2.5–10}$ and PM_{10}) are considered as dependent variables, while meteorological factors are considered as independent variables. The statistical significances of constant and coefficients are calculated by using SPSS 10.0.

2.2.5. Quality control

Blank test background contamination was monitored by using operational blanks (unexposed projection film and quartz filter) was processed simultaneously with field samples. The field blanks were exposed in the field when the field sampling box was opened to remove and replace field samples. Background contamination of heavy metals was accounted for by subtracting field blank values from the concentrations. Field blank values were very low, usually, below or around the method detection limits. In this study, the background contamination is insignificant and can be ignored. The results of the blank test are 0.41, 0.38, 0.32, 0.22, 0.25, 0.20 and 0.18 μg for Fe, Zn, Mn, Cu, Pb, Cr and Mg, respectively.

2.2.6. Recovery efficiency test

At least 10% of the samples are analyzed in spiking with a known amount of metal to calculate recovery efficiencies. The analysis procedure for the recovery test is the same as that described for the field samples. The recovery tests of metallic elements were 98, 96, 94, 106, 101, 104 and 102% for Fe, Zn, Mn, Cu, Pb, Cr and Mg, respectively. The range of recovery efficiency test varies between 94% and 106% and the relative standard deviation is smaller than 6%.

2.2.7. Reproducibility test

Repeat the analysis of the same standard solution for many times. The reproducibility test can be displayed the stability of instruments. The relative standard deviation varied between 98% and 101%, the mean relative deviation is smaller than 5%.

2.2.8. Detection limit

Detection limit was used to determine the lowest concentration level that can be detected to be statistically different from a blank. Method detection limit (MDL) was determined from selective the concentration slightly higher than the lowest concentration of the standard line. The procedure of repeat the lowest concentration of standard solution for twelve times was also used to estimate the standard deviation (S.D.). Method detection limit for metallic elements were 0.024, 0.008, 0.024, 0.013, 0.016, 0.011and 0.017 mg/l^{-1} for Fe, Mg, Cr, Cu, Zn, Mn and Pb, respectively. Method detection limit was determined from selected the concentration slightly higher than the low con-

Table 1
Sampling information and different particle size concentration in March 2004 to February 2005

Season	Month	TSP (μg m^{-3})	$PM_{2.5}$ (μg m^{-3})	$PM_{2.5–10}$ (μg m^{-3})	Temperature (°C)	Relative humidity (%)	Wind speed (ms^{-1})	Wind direction
(A) Taichung Harbor								
Spring	March	186.55	54.79	39.03	16.50	86	13.6	NNE
	April	171.25	52.89	38.26	19.10	83	9.8	NNW
	May	160.63	44.79	34.10	21.70	84	8.6	NNW
Summer	June	143.55	32.40	27.44	22.50	84	8.7	NNE
	July	121.84	47.05	18.14	22.60	86	7.2	NE
	August	118.32	33.02	15.55	22.80	87	7.8	SWW
Autumn	September	144.34	39.76	22.44	21.80	88	8.2	SWW
	October	157.56	36.92	20.45	19.00	79	8.5	NNE
	November	163.95	49.63	29.35	18.50	78	8.3	NE
Winter	December	173.75	52.31	39.93	15.60	77	9.7	NW
	January	175.61	46.02	38.22	13.50	78	10.4	NNW
	February	170.43	42.56	35.42	14.32	76	8.6	NW
	Average	157.31	44.35	29.86	18.99	82.17	9.12	NNW
	S.D.	21.98	7.94	9.13	3.18	4.03	1.74	
(B) WuChi Traffic area								
Spring	March	156.56	45.40	28.83	18.9	72	10.5	NNE
	April	132.92	42.47	29.12	21.6	75	8.7	NW
	May	100.79	41.39	23.87	24.4	76	6.9	NNW
Summer	June	84.70	38.92	28.49	27.5	70	6.0	NNE
	July	71.13	32.34	20.44	29.7	76	4.3	NE
	August	76.14	39.03	20.37	27.2	78	5.5	SWW
Autumn	September	79.55	28.16	13.68	27.2	73	6.5	SW
	October	102.03	31.89	15.56	24.2	72	6.9	NNE
	November	118.58	38.70	19.88	22.0	69	6.0	NE
Winter	December	140.86	36.25	24.54	17.2	77	6.4	NNW
	January	149.17	35.08	23.35	14.6	74	7.6	NNW
	February	138.51	32.84	21.72	16.4	76	6.8	NNW
	Average	112.58	36.87	22.49	22.57	73.98	6.83	NNW
	S.D.	31.03	5.10	5.18	4.76	2.99	1.66	

centration of the standard line, repeated this concentration for twelve times to estimate the standard deviation (S.D.). The MDL was equally to be (3 × S.D.).

3. Results and discussion

3.1. Sampling information

Table 1 shows the sampling information which includes the sampling season, sampling month, temperature (°C), relative humidity (%), wind speed ($m\,s^{-1}$) and wind direction at the TH and WT sampling sites of central Taiwan during four season period of March 2004 to February 2005. Simultaneous TH and WT samples were part in 40 groups be collected from March 2004 to February 2005. Air sample of two sampling sites (TH and WT) was collected for 24 h at the same time. The average sampling temperature was 18.99 and 22.57 °C during this sampling period at TH and WT sampling sites, respectively. The average relative humidity and wind speed at the TH sampling period was 82.17 °C and 9.2 $m\,s^{-1}$, respectively. The average relative humidity and wind speed at the WT sampling period was 73.98 °C and 6.83 $m\,s^{-1}$, respectively. In general, 24 h sampling period was performed in this study. TH sampling site has higher humidity and wind speed. The dominant wind direction was NNW at both sampling sites.

3.2. Ambient particulate mass concentration

Table 1 indicated the average concentration with different particle size (TSP, $PM_{2.5–10}$ and $PM_{2.5}$) during sampling period in central Taiwan. The result indicated the average total suspended particulate concentration in 1 year was 157.31 and 112.58 $\mu g\,m^{-3}$ at TH and WT sampling site, respectively. The average $PM_{2.5–10}$ and $PM_{2.5}$ concentration was 29.86 and 44.35 $\mu g\,m^{-3}$ at the TH sampling site. In addition, the average $PM_{2.5–10}$ and $PM_{2.5}$ concentration was 22.49 and 36.87 $\mu g\,m^{-3}$ at the WT sampling site. Fine particle ($PM_{2.5}$) size was the dominant species at TH and WT sampling site.In general, winter has the highest average TSP, $PM_{2.5}$, $PM_{2.5–10}$ concentrations among four seasons at Taichung Harbor sampling site. Though spring has highest average TSP concentration, it only reflect the regular dust storm occurred season in spring during sampling period. Thus, March is, apparently, has the highest TSP, $PM_{2.5}$, $PM_{2.5–10}$ concentrations. However, winter still owned the high average TSP concentrations among four seasons at TH sampling site if ambient air particulate concentrations were excluded from sparing.

The average particulate concentrations ratios of TSP in spring to summer, autumn and winter were 1.35, 1.11 and 1, respectively. And average particulate concentrations ratios of $PM_{2.5–10}$ in spring to summer, autumn and winter were 1.82, 1.54 and 1.08, respectively. In addition, average particulate concentrations ratios of $PM_{2.5}$ in spring to summer, autumn and winter were 1.36, 1.21 and 1.08, respectively at TH sampling site.

As for WT sampling site, the average particulate concentrations ratios of TSP in spring to summer, autumn and winter were 1.68, 1.3 and 0.91, respectively. And average particulate concentrations ratios of $PM_{2.5–10}$ in spring to summer, autumn and winter were 1.17, 1.66 and 1.17, respectively. In addition, average particulate concentrations ratios of $PM_{2.5}$ in spring to summer, autumn and winter were 1.17, 1.31 and 1.24, respectively at the WuChi Traffic (WT) sampling site.

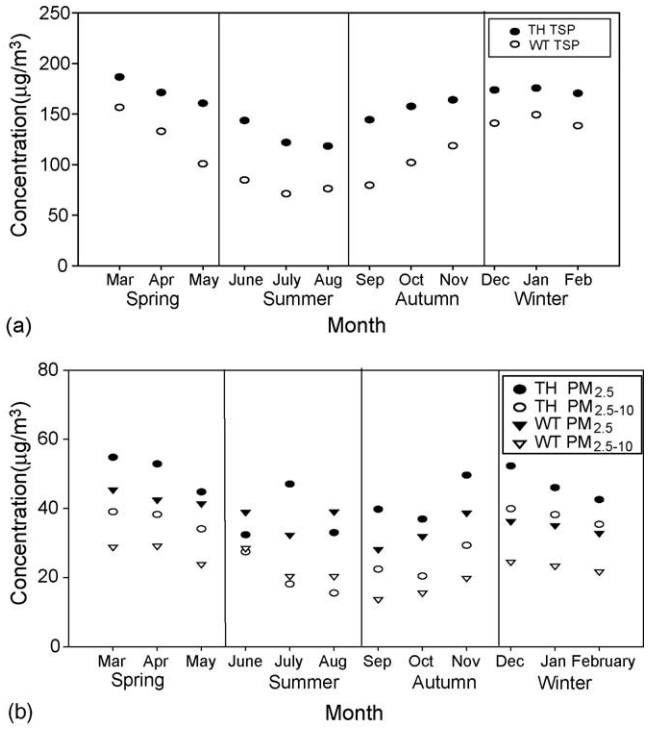

Fig. 2. (a) TSP average concentration in TH and WT sampling site among four seasons. (b) $PM_{2.5–10}$ and $PM_{2.5}$ average concentration in TH and WT sampling site among four seasons.

Fig. 2a and b showed the seasonal variation of TSP and different particulate size concentrations during period of March 2004 to February 2005 at TH and WT sampling sites. In Fig. 2a, it displayed the period variation of TSP concentrations, like Cosine function, during 1 year. The lowest and highest particulate concentrations for TSP were occurred in summer season (August) and spring season (March), respectively. The results also indicated TSP concentrations have the same distribution tendency among four seasons in either TH or WT sampling site. Fig. 2b indicated that the concentrations of $PM_{2.5}$ were almost greater than $PM_{2.5–10}$, but the variations among four seasons were not regular. The variation of the particulate concentrations at TH sampling site was also greater than WT sampling site among four seasons. However, these variations values were not significant among four seasons.

Comparing with the previous study [13,14], air samples collected from Beirut, THU, JES, and SES four sampling sites revealed that the average TSP concentrations were 1.06, 1.62, 1.71, and 2.10 times to Taichung Habor, respectively. In addition, the average TSP concentrations were 1.49, 2.27, 2.39, and 2.94 times to WuChi Traffic, respectively. In above regions, the vehicle-induced emissions, moment of motor vehicles on dusty road and on-going construction activities are the major potential sources for particulates. Based on some international standards limit values (European Union, EU; United States Environmental

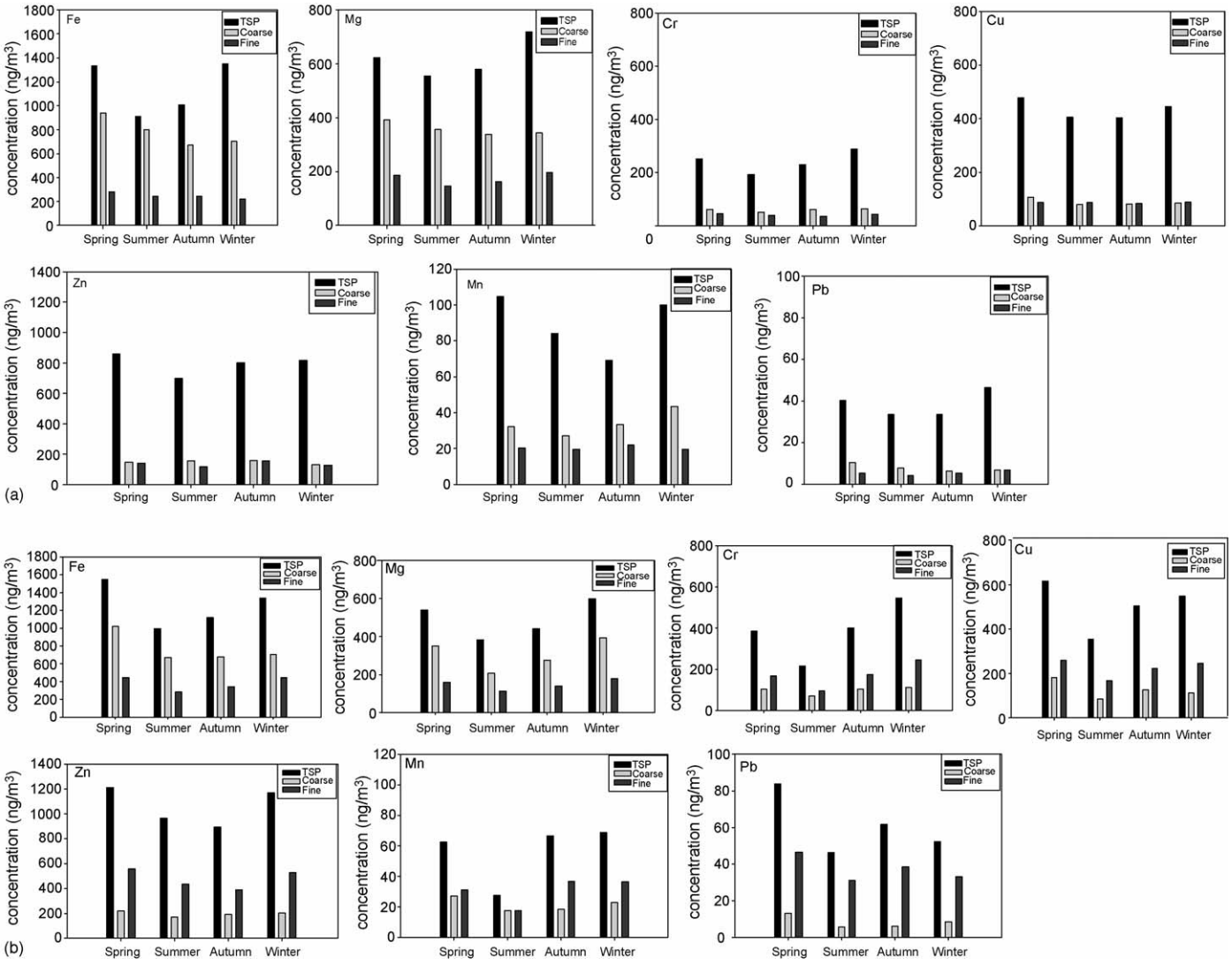

Fig. 3. (a) Different particle sizes concentration of metallic elements (Fe, Mg, Pb, Zn, Cr, Mn and Cu) at TH sampling site during four seasons. (b) Different particle sizes concentration of metallic elements (Fe, Mg, Pb, Zn, Cr, Mn and Cu) at WT sampling site during four seasons.

Protection Agency, USEPA; World Health Organization, WHO), the values of the measured TSP concentrations were higher than the air quality standards, and the measured concentrations of the TSP particulates were used to investigate the human health studies.

3.3. Ambient airborne particle concentration of metallic metals

The average metallic elements (Fe, Mg, Pb, Zn, Cr, Mn and Cu) concentrations were measured with different particle size of ambient suspended particle during sampling period at TH sampling sites in central Taiwan in Fig. 3a. Metallic element of Fe and Mg indicated concentration for various particle size range were $TSP > PM_{2.5–10} > PM_{2.5}$ during sampling period and the metallic elements of Fe and Mg have higher concentration compared with other metallic elements (Pb, Zn, Cr, Mn and Cu). The higher and lower concentration (TSP and $PM_{2.5–10}$) of metallic element Fe was occurred in autumn and summer season, respectively. In addition, the higher and lower concentration (TSP) of metallic element Mg was occurred in winter and summer season however, the higher and lower concentration ($PM_{2.5–10}$) of metallic element Mg was occurred in spring and autumn season, respectively. However, this phenomenon is not significant in PM2.5 particle size. Metallic elements of Pb, Zn, Cr, Mn were indicated concentration for various particle size range were $TSP > PM_{2.5–10} > PM_{2.5}$ during sampling period. The metallic element of Cr and Pb average TSP, $PM_{2.5–10}$ and $PM_{2.5}$ concentration in summer season has occurred lowest concentration. Besides, the higher TSP, $PM_{2.5–10}$ and $PM_{2.5}$ concentration of Cr was occurred in winter season. In addition, metallic element of Cu higher average concentrations in particle size TSP and $PM_{2.5–10}$ was occurred in spring season. But in $PM_{2.5}$ particle size, this phenomenon is not significant in $PM_{2.5}$ particle size in metallic element Cu was in among four seasons.

Fig. 3b shows the average metallic elements (Fe, Mg, Pb, Zn, Cr, Mn and Cu) concentration with different particle size of ambient suspended particle during sampling period at WT

Table 2
Correlation coefficients of total, coarse ($PM_{2.5–10}$) and fine ($PM_{2.5}$) particulates during sampling period at TH and WT sampling sites

TH-TSP

Species	Fe	Mg	Pb	Zn	Cr	Mn	Cu
Fe	1.00						
Mg	0.30	1.00					
Pb	0.13	0.23	1.00				
Zn	0.76**	0.17	0.41	1.00			
Cr	0.31	0.32	0.25	0.33	1.00		
Mn	0.36	0.42	0.17	0.16	0.53	1.00	
Cu	0.30	0.37	0.04	0.47	0.13	0.34	1.00

WT-TSP

Species	Fe	Mg	Pb	Zn	Cr	Mn	Cu
Fe	1.00						
Mg	0.54	1.00					
Pb	0.22	0.14	1.00				
Zn	0.76**	0.42	0.64	1.00			
Cr	0.32	0.60	0.59	0.21	1.00		
Mn	0.70	0.31	0.24	0.33	0.23	1.00	
Cu	0.31	-0.18	0.55	0.16	0.37	0.53	1.00

TH-$PM_{2.5–10}$

Species	Fe	Mg	Pb	Zn	Cr	Mn	Cu
Fe	1.00						
Mg	0.73**	1.00					
Pb	0.35	0.41	1.00				
Zn	0.58	0.35	0.32	1.00			
Cr	0.31	0.50	0.26	0.21	1.00		
Mn	0.76**	0.71	0.31	0.12	0.61	1.00	
Cu	0.82**	0.32	0.01	0.38	0.12	0.49	1.00

WT-$PM_{2.5–10}$

Species	Fe	Mg	Pb	Zn	Cr	Mn	Cu
Fe	1.00						
Mg	0.26	1.00					
Pb	0.38	0.18	1.00				
Zn	0.42	0.47	0.18	1.00			
Cr	0.25	0.67	0.63	0.68	1.00		
Mn	0.41	0.26	0.27	0.31	0.16	1.00	
Cu	0.32	0.88**	0.65	0.49	0.72	0.62	1.00

TH-$PM_{2.5}$

Species	Fe	Mg	Pb	Zn	Cr	Mn	Cu
Fe	1.00						
Mg	0.73**	1.00					
Pb	0.64	0.33	1.00				
Zn	0.48	0.16	0.24	1.00			
Cr	0.25	0.69	0.36	0.31	1.00		
Mn	0.37	0.65	0.28	0.18	0.35	1.00	
Cu	0.26	0.32	0.14	0.51	0.18	0.37	1.00

WT-$PM_{2.5}$

Species	Fe	Mg	Pb	Zn	Cr	Mn	Cu
Fe	1.00						
Mg	0.82**	1.00					
Pb	0.18	0.18	1.00				
Zn	0.67	0.71	0.29	1.00			
Cr	0.52	0.52	0.54	0.61	1.00		
Mn	0.26	0.29	0.18	0.22	0.33	1.00	
Cu	0.51	0.76**	0.41	0.61	0.68	0.15	1.00

** Correlation is significant at the 0.01 level (two-tailed).

sampling site in central Taiwan. The metallic elements of Fe and Zn have higher concentration compared with other metallic elements (Pb, Mg, Cr, Mn and Cu). Metallic elements of Fe and Mg indicated concentration for various particle size range were TSP > $PM_{2.5–10}$ > $PM_{2.5}$; however, metallic elements of Zn, Mn, Cr, Pb and Cu indicated concentration for various particle size range were TSP > $PM_{2.5}$ > $PM_{2.5–10}$ during sampling period.

3.4. Ambient airborne particle correlation coefficients of metallic metals

Table 2 shows the correlation coefficients matrix of eight selective metallic elements with different particle size (TSP, $PM_{2.5–10}$ and $PM_{2.5}$) during sampling periods at TH and WT sampling site. In TH sampling site, higher correlation coefficient was observed on total suspended particulates of metallic elements Fe and Zn. And in WT sampling site, higher correlation coefficients displayed on total suspended particulates of metallic elements Fe and Zn, Fe and Mn. On coarse particulates, higher correlation coefficients were observed among Fe, Mg, Mn and Cu at TH sampling site. In addition, higher correlation coefficients were observed among Cu, Mg and Cr at WT sampling site. These results indicated that the higher correlation coefficients with metallic elements might come from the same pollutant sources sampling period.

The correlation coefficients matrix on fine particulate, higher correlation coefficients were also observed only on Fe and Mg at TH sampling site. Higher correlation coefficients were observed among Fe, Mg, Zn and Cu at WT sampling site. These results indicated that the species of metallic element correlation coefficient was changed by different particle size. Compared with coarse and fine particulates, the same correlation coefficients were only occurred between Fe and Mg at TH. In WT, compared with coarse and fine particulates displayed the metallic element of Mg and Cu has higher correlation coefficients.

3.5. Ambient airborne particle principal component analysis of metallic metals

Metallic element profiles for sources of relevance in other studies were listed in Table 3a. The metallic element profiles for stationary sources were included that the soil, suspended dust, vehicular emission, industrial process, incinerator, sea salt, coal combustion, metal industry, oil combustion and construction sites [15–26,28]. The following metallic elements have been identified as markers for various sources in atmosphere: soil and resuspended dust—Ca, Mg, Al, Si, Fe and Mn; vehicular emission—Cr, Pb, Cu, Zn, Cd, Sb, Br, Fe and Ba; industrial process—Mn, Zn and K; incinerator—K, Zn and Pb; sea salt—Na, Ca, Mg and K; coal combustion—Cr;

Table 3a
Indicatory metallic elements for various major sources

Predominant species	Source	Reference
Ca, Mg, Al, Si, Fe and Mn	Soil and resuspended dust	[15,18–20,23,25,28]
Cr, Pb, Cu, Zn, Cd, Sb, Br, Fe and Ba	Vehicular emission	[18–20,23–26]
Mn, Zn and K	Industrial process	[15,19]
K, Zn and Pb	Incinerator	[22,24]
Na, Ca, Mg, and K	Sea salt	[16,25]
Cr	Coal combustion	[21]
Fe, Mn and Pb	Metal industry	[22,24]
Ni and V	Oil combustion	[17,20]
Al, Fe, Si, Mn and Ti	Construction site	[20]

metal industry—Fe, Mn and Pb; oil combustion—Ni and V; construction—Al, Fe, Si, Mn and Ti.

Principal component analysis (PCA) with Varimax rotation and retention of principal components having eigenvalues >1 (SPSS 10.0) was used to identify the possible pollutant sources [27]. Table 3b shows the factor analysis on total, coarse and fine particulate metallic element concentrations during sampling period at TH and WT sampling sites. The PCA results showed that two and three factors explain the main part of the data variance, therefore one element for each factor has been chosen as a tracer.

At the WT sampling site, factor 1 explained 53.74% of the total variance of the data and had high loading for Zn (0.86) and Cu (0.85), which were identified as industrial/traffic emission sources. More factories located near WT area, possessed large traffic volume especially during work time; this was the main reason for the above mentioned results. Factor 2 is relative to soil dust or resuspended source with high loading of Fe (0.88) and Mn (0.72). At the TH sampling site, 50.81% of the total variance of the data was observed in factor 1. Higher loading of Fe (0.86), Zn (0.79), Pb (0.76), and Mn (0.68) were contributed by traffic emission and the soil source. Factor 2 had high loading of Mg

Table 3b
Factor analysis of TSP, coarse ($PM_{2.5–10}$) and fine ($PM_{2.5}$) particulates metallic element concentrations data at TH and WT sampling sites

	Variables							
	TSP			$PM_{2.5–10}$		$PM_{2.5}$		
	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 1	Factor 2	Factor 3
TH								
Fe	0.86	0.23	–	0.03	0.89	0.87	–	0.01
Mg	0.61	0.86	–	0.92	0.13	0.91	0.09	0.12
Pb	0.76	–	–	0.78	–	0.08	0.92	0.06
Zn	0.79	0.18	–	0.71	0.16	0.84	0.12	0.3 1
Cr	0.45	0.20	0.08	0.62	0.62	0.61	–	0.38
Mn	0.68	0.51	0.27	0.15	0.72	0.10	–	0.94
Cu	–	–	0.88	0.92	0.18	0.85	–	0.01
Eigenvalue	8.84	1.37	1.15	3.58	1.76	3.82	1.41	1.06
Proportion of variance (%)	50.81	18.92	12.67	50.82	25.38	54.91	18.25	12.86
Cumulative %	50.81	69.74	82.41	50.82	76.2	54.91	73.16	86.02
Origin	Traffic soil	Marine salt	Smelter	Marine salt traffic	Dust	Dust traffic	Anthropogenic	Crustal

	Variables					
	TSP		$PM_{2.5–10}$		$PM_{2.5}$	
	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2
WT						
Fe	0.42	0.88	0.95	0.19	0.82	0.35
Mg	0.31	0.38	0.86	0.05	0.91	0.09
Pb	0.56	0.21	–	0.76	–	–
Zn	0.86	0.12	0.38	0.71	0.32	0.18
Cr	0.32	-0.2	0.51	0.73	0.36	0.81
Mn	0.41	0.72	0.91	0.28	0.72	0.31
Cu	0.85	0.11	0.82	0.15	0.78	0.24
Eigenvalue	2.64	1.16	4.1	1.37	3.613	1.109
Proportion of variance (%)	53.74	19.63	58.63	19.61	51.62	15.76
Cumulative %	53.74	73.37	58.63	78.24	51.62	67.38
Origin	Traffic industrial	Soil resuspended	Dust anthropogenic	Coal combustion and incinerator	Dust anthropogenic	Unknown

(0.86); this phenomenon indicates a marine salt source. Marine salt from the Taiwan Strait, about 100 m from the TH sampling site, was the main cause of this result. High loading of Cu (0.88) for factor 3b indicated that emissions of smelter industry from metallurgical procedure were also observed in this area.

In TH sampling site with fine particle size, factor 1 explained 54.91% of the total variance of the data and had high loadings for Fe (0.87), Mg (0.91), Pb(0.78), Cu (0.85), and Zn (0.84), which were identified as dust and traffic emission sources on fine particulates [23]. Higher factor loading of Pb (0.92) and Mn (0.94) were observed on factor 2 and factor 3 for fine particulates. The contributors of Pb were come from anthropogenic activities but not for traffic emission [17,22]. As for Mn, high factor loading values were observed simultaneously. Besides, the major contributors of Mn are anthropogenic and natural sources [19,20]. According to the above reasons, crustal source was the third major pollutant source affects the fine particulates concentrations at TH sampling site.

As for coarse particulates, high factor loadings of Mg (0.92), Cu (0.92) and Zn (0.71) were observed in TH sampling site. This suggested that the traffic vehicle emissions and marine salt were the major contributors [18,23]. Factor 2 showed high factor loadings of Fe (0.89) and Mn (0.72) on coarse particulates during sampling period. The soil dust was responsible for these results. This finding is similar to previous study by Funasaka et al. [24].

From Table 3b, factor 1 showed 51.62% of the total variance of the data and high factor loading of Fe (0.82) and Mg (0.91) were found on fine particulates at the WT sampling site. Thus, soil was inevitably became the major pollutant source. Besides, higher factor of Cu (0.78) and Mn (0.72) were also observed in factor 1 at WT sampling site. Salvador et al. [25] indicated Cu and Zn were associated with type wearing. In addition, Zn has been suggested as a good marker of gasoline engine emissions. Based on above reasons, it suggested anthropogenic activities such as industrial process and traffic emission were the major pollutant sources in fine particulates. Chromium had high factor loading in fine particulate in factor 2. But no obvious contributor was observed according to the limit information.

The same phenomena were occurred on coarse particulate values at WT sampling site. The descriptions of coarse particulates were the same to fine particulates. High factor loading of Cr (0.73), Zn (0.71) and Pb (0.76) were observed on factor 2 in coarse particulates. Chromium and zinc were enriched from anthropogenic activities such as coal combustion and incinerator emissions [21,22]. The distance of Taichung Thermal Power Plant which located at the west side of the WT sampling site is about 10 km. Hence, the influence of Taichung Thermal Power Plant seems important on coarse particulate at WT sampling site.

3.6. Correlation between ambient airborne particles and meteorological parameters

In this section, a non-parametric (Spearman) correlation analysis is taken to investigate the ambient airborne particles (TSP, $PM_{2.5}$ and $PM_{2.5–10}$) correlate with the meteorology data at TH and WT sampling sites. The results of the correlation analysis of the particulate matters (TSP, $PM_{2.5}$ and $PM_{2.5–10}$) and the meteorological parameters (temperature, wind speed, relative humidity and wind direction) are presented in the summary Tables 4a and 4b. In Tables 4a and 4b we can see that the particulate concentrations TSP has higher correlation coefficients with wind speed and inversely correlated with temperature at TH sampling sites. This phenomenon is also observed at WT sampling site. The correlation coefficients between the other particulate

Table 4a
Spearman rank correlation coefficients of particulate concentrations and meteorological factor at TH sampling site

	TSP	$PM_{2.5–10}$	$PM_{2.5}$	Temperature	Relative humidity	Wind speed	Wind direction
TSP	1.0						
$PM_{2.5–10}$	0.930**	1.0					
$PM_{2.5}$	0.727**	0.713**	1.0				
Temperature	−0.881**	−0.755**	−0.462	1.0			
Relative humidity	−0.545	−0.527	−0.228	0.784**	1.0		
Wind speed	0.855**	0.872**	0.480	−0.658*	−0.386	1.0	
Wind direction	−0.021	0.075	−0.271	−0.014	0.090	−0.016	1.0

* Correlation is significant at the 0.05 level (two-tailed).
** Correlation is significant at the 0.01 level (two-tailed).

Table 4b
Spearman rank correlation coefficients of particulate concentrations and meteorological factor at WT sampling site

	TSP	$PM_{2.5–10}$	$PM_{2.5}$	Temperature	Relative humidity	Wind speed	Wind direction
TSP	1.0						
$PM_{2.5–10}$	0.497	1.0					
$PM_{2.5}$	0.301	0.748**	1.0				
Temperature	−0.928**	−0.305	−0.126	1.0			
Relative humidity	0.162	0.074	0.028	−0.049	1.0		
Wind speed	0.723**	0.446	0.333	−0.633*	−0.214	1.0	
Wind direction	−0.047	−0.014	0.032	−0.133	0.496	0.205	1.0

* Correlation is significant at the 0.05 level (2-tailed).
** Correlation is significant at the 0.01 level (2-tailed).

Table 5
Regression coefficients with the different particulate size concentrations

Particulate size	Regression coefficients						
	α	β_1	β_2	β_3	β_4	n	R^2
TSP (TH)	177.87	−8.79	5.23	3.42	0.92	1	0.935
$PM_{2.5–10}$ (TH)	17.02	0.27	1.34	3.21	−0.57	2	0.915
$PM_{2.5}$ (TH)	37.52	0.77	0.45	4.13	−1.08	4	0.664
TSP (WT)	109.45	−4.22	−0.56	6.68	2.07	1	0.982
$PM_{2.5–10}$ (WT)	16.66	0.62	0.33	5.63	−1.14	2	0.772
$PM_{2.5}$ (WT)	33.47	1.39	−0.04	4.00	−1.43	4	0.613

concentrations and the meteorological parameters are not significant.

According to the above spearman rank, the major parameter wind speed and temperature are used to establish the regression equation obtained for TSP, $PM_{2.5}$ and $PM_{2.5–10}$ at TH and WT sampling sites. According to the above meteorological parameters in Table 1, the characteristic function F of the particulate concentrations could be expressed as

$$F = f(\text{wind speed},\ \text{temperature},\ \gamma_i) \quad (1)$$

The regression equations obtain for TSP, $PM_{2.5}$ and $PM_{2.5–10}$ concentrations are expressed as

$$F = \alpha_1 + [\beta_1(\text{wind speed}) + \beta_2(\text{temperature})] \times \cos[\beta_3(\gamma_i/12) + \beta_4]^n \quad (2)$$

where F, particulate concentrations ($\mu g\ m^{-3}$); temperature, temperature (°C); wind speed, wind speed (m/s); γ_i, is the sequence of the month in 1 year, α_1, β_1, β_2, β_3 and n are the regression coefficients. The results of the regression coefficients are shown in Table 5. From Table 5, we can see that the values of the correlation coefficients (R^2) between the TSP, $PM_{2.5}$ and $PM_{2.5–10}$ particulates concentrations and meteorological parameters are form 0.613 to 0.982. Eq. (2) presents the prediction equation of the TSP, $PM_{2.5}$ and $PM_{2.5–10}$ particulates concentrations. The regression coefficient α_1 means the mean value of the particulates concentrations. The coefficients β_1 (wind speed) and β_2 (temperature) mean the secondary variation amplitude with the triangle function of cosine in Eq. (2). By taking out the mean values, the above equation reveals that the TSP, $PM_{2.5}$ and $PM_{2.5–10}$ concentrations vary with different wind speed, temperature, and different months. The above regression equations reveal that the concentrations of TSP has higher correlation coefficients (R^2) than $PM_{2.5}$ and $PM_{2.5–10}$. The variations of the tendency are like the triangle function of Cosine including the parameter γ_i at TH and WT sampling sites. The fine particulate concentration $PM_{2.5}$ presents the random tendency during the sampling period at TH and WT sampling sites.

4. Conclusions

Results of measurements presented in this paper five the following conclusions:

(1) The average total suspended particulate concentration in 1 year was 157.31 and 112.58 $\mu g\ m^{-3}$ at TH and WT sampling site, respectively. The average $PM_{2.5–10}$ and $PM_{2.5}$ concentration was 29.86 and 44.35 $\mu g\ m^{-3}$ at the TH sampling site. In addition, the average $PM_{2.5–10}$ and $PM_{2.5}$ concentration was 22.49 and 36.87 $\mu g\ m^{-3}$ at the WT sampling site. Fine particle ($PM_{2.5}$) size was the dominant species at TH and WT sampling site.
(2) The lowest and highest particulate concentration for TSP were occurred in summer season (August) and spring season (March), respectively. The concentrations of $PM_{2.5}$ were almost greater than $PM_{2.5–10}$, and the variations among four seasons were not regular and significant at TH and WT sampling site.
(3) The higher and lower concentration (TSP and $PM_{2.5–10}$) of metallic element Fe and Mg was occurred in winter and summer season, respectively. However, this phenomenon is not significant in $PM_{2.5–10}$ particle size.
(4) The results identified as dust, traffic, anthropogenic and crustal were the sources on fine particulates. And the results also indicated identified as marine salt and traffic were the sources on coarse particulates at Taichung Harbor near Taiwan Strait.
(5) The results identified as dust and anthropogenic were the sources on fine particulates. And the results also indicated identified as dust, anthropogenic, coal combustion and incinerator were the sources on coarse particulates at WT sampling site.
(6) According to the non-parametric (Spearman) correlation analysis, the particulate concentrations TSP has higher correlation coefficients with wind speed and inversely correlated with temperature at TH and WT sampling sites. TSP concentrations present the period variation, like cosine function, during 1 year.

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