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Ambient Trace Metals Sources in Taichung, Taiwan: Principal Component Analysis

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ABSTRACT

There were eight trace metals, including As, Hg, Mn, Fe, Zn, Cr, Cu, and Pb, measured in the total suspended particles collected in central Taiwan using PS-1 samplers at five different sites. Overall, these trace metal concentrations were lower in summer than other seasons, and higher in industrial areas than in wetland. Principal Component Analysis (PCA) was utilized to identify the pollutant sources for each site. Three distinct sources were found in this study, and the results suggest that mobile sources were the main factor contributing to Mn, Fe, Zn, and Cr emissions, which were mainly from urban areas and in the vicinity of highways intersections. Within addition, municipal solid waste incinerators are significantly associated with emissions of As, Hg, Cu, and Pb. Finally, smelters were associated with high Hg, As, Fe, and Zn emissions from local industrial areas. However, contributions from two known point sources, a coal-fired power plant and a steeling plant could not be distinguished from the PCA results. This may be due to the prevailing wind directions and limitations of the measurements used in this work. These two sources are located to the west of these sampling sites, while the prevailing wind direction in this area was southwest.

Keywords: Principal component analysis; Conditional probability function; Trace metals; Source identification; Central Taiwan.

INTRODUCTION

Most particulate trace metals have been considered to be the toxic pollutants (Harrison and Yin, 2000; Okuda *et al.*, 2004; Witt *et al.*, 2010a, b); however, mercury (Hg) and arsenic (As) also exist in gaseous phase due to their high vapor pressures. Long term exposure to high levels of PM can cause significant risk to human health (Ny *et al.*, 2011). Thus, understanding the level of PM_{10} and $PM_{2.5}$ (PM with an aerodynamic diameter less than 2.5 μm) in urban areas is important. Techniques such as source apportionment have been developed to control PM pollutants (Wang *et al.*, 2012). Particulate trace metal concentrations are attributed to their distinguished unique emission sources. In general, As, Cr, Cu, Mn, and Zn have been reported as the indicators of anthropogenic pollutants (Pacyna and Pacyna, 2001; Samara *et al.*, 2003). Copper industry and coal combustion are the primary sources of As (Manoli *et al.*, 2002; Tian *et al.*, 2010; Minguillòn *et al.*, 2012; Shi *et al.*, 2012). The main emission sources of Pb, Cu, Zn, Ni, and Cd are mobile sources, including gasoline and diesel vehicles (Pacyna and Pacyna, 2001). A previous study suggested that Pb, Cd, Ni, and Zn originate from a common local source (Gelado-Caballero *et al.*, 2012). Although Minguillòn *et al.* (2012) stated high Pb concentrations might be related with thermal processes, they also concluded a factor associated with Zn and Cu from road traffic. V and Ni are considered to be the markers of oil combustion (Rajšić *et al.*, 2008). Emission sources of Hg have been extensively investigated and classified into coal combustion, solid waste incinerators, steeling industries, mobile sources, and wildfires (Landis *et al.*, 2007; Choi *et al.*, 2008; Huang *et al.*, 2010;). The particulate matters containing these pollutants have been considered to be harmful for human via direct and/or indirect pathways (Adriano, 1986). For example, fine particles could easily be inhaled and thus transported to the respiratory system or even the lung, or deposited into ecosystems from the atmosphere, uptaken by organisms which results in bioaccumulation (Adriano, 1986). Some trace metals, including V, Cr, Mn, Fe, Ni, Cu, and Zn, are believed to introduce to lung damages when deposited in place (Claiborn *et al.*, 2002; Osonio-Vargas *et al.*, 2003). The major adverse health effect

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of Hg is nervous system damages and cardiovascular disease resulting from fish and rice consumption (Jewett and Duffy, 2007; Zhang *et al.*, 2010). The long-term exposure to inorganic As has been demonstrated to be linked to dermal cancers and blackfoot disease (Chang *et al.*, 2010).

There are two types of major data analyses used to explore the correlation between ambient air species and emissions sources: source and receptor models. The former one incorporates meteorological data, point source emission profiles, and chemical and physical reactions to attribute the contribution of certain point sources to the pollutant concentrations at a particular sampling site, such as Community Multiscale Air Quality Model (CMAQ) (Byun and Ching, 1999). However, the required inputs for this kind of model shows high uncertainty and a complicated calculate scheme is required for application (Byun and Ching, 1999). Nevertheless, a simple and low cost receptor model could be applied to identify the origins of plumes, such as principal component analysis (PCA), coupled with Hybrid Single Particle Lagrangian Integrated Trajectory Model (HSYPLIT) and/or conditional probability function (CPF) (Rajšić *et al.*, 2008; Huang *et al.*, 2010; Cheng *et al.*, 2012). For example, Rajšić *et al.* (2008) identified that the particulate matter observed high of Zn, Mn, Fe, and Al was from road dust, while V and Ni were emitted from oil combustion, and Cu, Cd, and Pb were linked to mobile sources by applying PCA to indentify emission sources of trace metals.

In this study, total suspended particles were collected at five different sites in central Taiwan, including suburban/coastal, urban, industrial, residential, and wetland areas for a year. Previous studies at these sites focused on the comparison of dry deposition velocities of trace metals from model calculation and surrogate surfaces measurements (Fang *et al.*, 2012; Zhang *et al.*, 2012). Additionally, meteorological data, including air temperature, relative humidity, wind speed, and wind direction, were also measured at individual sites. PCA and CPF were used to identify potential emission sources of trace metals based on measurements conducted on those samples collected.

METHODS

Sampling Sites

Five sites in central Taiwan were selected to investigate the behaviors of trace metals and their corresponding emission sources in various areas, including suburban/coastal (Beshi, BS), urban (Changhua, CH), industrial (Quanxing, QX), residential (Hemei, HM), and wetland areas (Gaomei, GM) (Fig. 1). The significant potential trace metals emission point sources in this region include a coal-fired power plant (CFPP), four municipal solid waste incinerators (MSWIs), a smelting manufacture, and several heavy-density industrial regions. Besides for these point emission sources, trace metals concentrations could be originated from mobile sources (von Schneidemesser *et al.*, 2010), especially in central Taiwan where heavy traffic occurs frequently. Details of these sampling sites have been elaborated by Fang *et al.* (2012) and Zhang *et al.* (2012).

Sampling Methods

High volume samplers (PS-1, Graseby-Anderson, General Metal Work) were used to collect ambient air total suspended particles (TSP) at a flow rate of approximately 200 LPM using quartz fiber filters (diameter 10.2 cm) for 24 hours. The filters were stored in a sealed CD box in a freezer before sampling. Pre-weight and sample weight were recorded before and after individual sampling. The detail operations and procedures for the TSP measurements were elaborated in detail in previous studies (Fang *et al.*, 2009; Fang *et al.*, 2012).

Chemical Analysis

Metallic Elements Analysis

One quarter of each collected filters were cut out for the digestion process, which were then cut into tiny pieces and

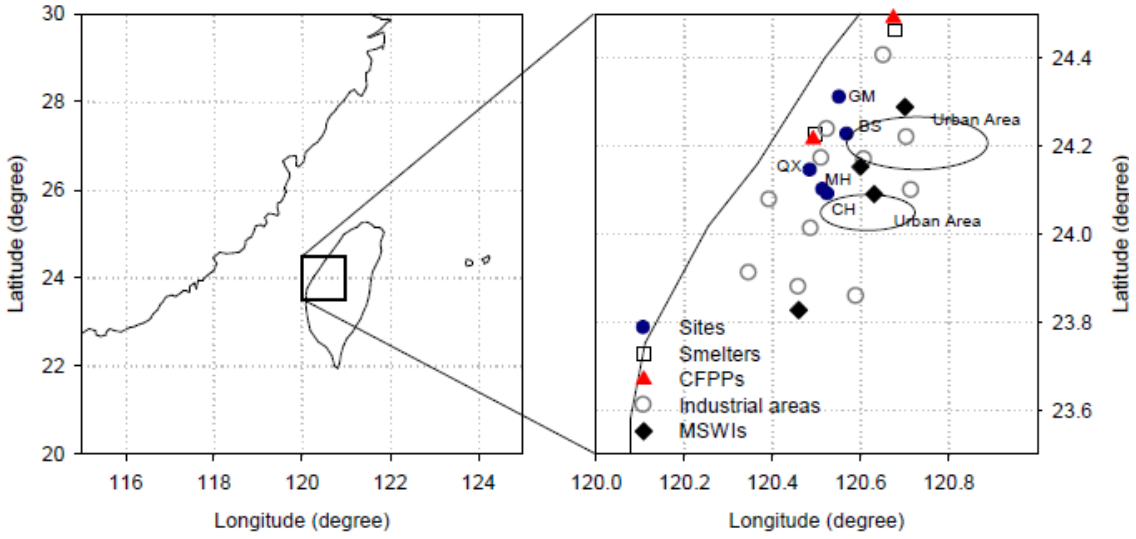

Fig. 1. Summary of the sampling sites. Blue close circles, black open squares, red close triangles, grey open circles, and black diamonds represent the locations of sampling sites, smelters, power plants, industrial areas, and MSWIs.

placed in a Teflon container. Next, 3 mL of hydrochloric acid (HCl) and 9 mL of nitrate (HNO_3) were mixed together and then poured into the Teflon container. The samples were then incubated under 50°C on a heated hotplate for 2 hours. After digestion on the hotplate, the samples were filtered out. Following filtration, the sample solution was added with 0.2% HNO_3 to yield a solution of final volume 100 mL (Taiwan EPA, 2006). The sample solution mixtures were stored at 4°C refrigerator before inductively coupled plasma-atomic emission spectrometer (ICP-AES) analysis. Concentrations of Mn, Zn, Cr, Cu, and Pb were determined using ICP-AES. ICP-AES analysis was carried out using a PerkinElmer Optima 2100 Plasma Emission Spectrometer. A 30 seconds delay and an argon gas plasma flow rate of 15 Lpmn were utilized. The nebulizer and sample flow rates were set at 0.65 Lpm and 1.5 Lpm, respectively (Fang *et al.*, 2012). As for metallic element As analysis, the digestion procedure was the same as the other metallic elements analysis described above. However, after digestion procedure, sample solutions for As measurements were analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Perkin Elmer Sciex ELAN DRCII) (Fang *et al.*, 2011, 2012).

Hg Analysis

Concentrations of Hg from each sample (quartz filter) were analyzed by a direct Hg analyzer (DMA-80 Milestone, Inc., Shelton, CT, USA). Approximately 30 mg of the filter sample was loaded directly into the DMA and analyzed using methods described in detail previously (Basu and Dietz, 2009). The methodology is based on a thermal decomposition of the sample and collection of the Hg vapor on a gold amalgamator. The filter samples were placed into a sampling boat and transferred to a combustion tube containing a catalyst. The sample is first dried at 200°C prior to combustion at 615–650°C in an oxygen atmosphere. The Hg vapor is collected in a gold amalgamator and after a predefined time at 170°C the gold amalgamator is heated up to 900°C. The released Hg is transported to a heated curette at 125°C and then analyzed by atomic absorption spectrometry (AAS) using a silicon UV diode detector. The operation conditions were: drying for 30 seconds at 200°C, decomposition for 90 seconds at 650°C, and combustion for 90 seconds at 650°C.

Quality Control

Metallic Elements Quality Control

Background contamination contribution was monitored using operational field blanks (unexposed quartz filter), which were processed simultaneously during sample collections. The field blanks were exposed in the field when the field sampling box was opened to change out samples. Background contamination of heavy metals was accounted for by subtracting field blank values from the concentrations. Field blank values were usually below or around the method detection limits (MDLs). In this study, the background contamination was insignificant and negligible. Background results of the one quarter cut of field blank were 0.31, 0.38, 0.35, 0.22, 0.20, 0.21and 0.19 μg of Mn, Fe, Zn, Cr, Cu, Pb and As, respectively. The field blanks for metallic elements were generally no more than 5% of those from collected exposed samples. At least 10% of the samples were analyzed by spiking with known amount of metal to calculate recovery efficiencies. The individual recoveries of metallic elements were Mn (92%), Fe (101%), Zn (97%), Cr (108%), Cu (104%), and Pb (103%).

Hg Quality Control

Analytical accuracy and precision were determined by using Standard Reference Materials (DOLT-3 and TORT-2 from National Research Council of Canada) and analyses of duplicate samples(Fang *et al.*, 2010). Average recovery rates of DOLT-3 and TORT-2 for total mercury (Hg) were within the certified values (± 10%). The detection limit for the direct Hg analyzer (DMA-80 Milestone, Inc., Shelton, Connecticut, USA) was 0.022 ng. And all the quality control field blanks were all less than 0.022 ng Hg.

Analytical Methods

Principal Component Analysis (PCA)

PCA is a statistical tool to reduce large number of variables to small number of factors that describe the principal variability or joint behavior of that data set (Jackson, 1991). Geometrically, this new set of variables represents a principal axis rotation of the original coordinate axes of the variables around their mean (Jackson, 1991). Trace metals concentrations in TSP, including As, Hg, Mn, Fe, Zn, Cr, Cu, and Pb, were examined by PCA. However, because of the different scale among individual metal concentrations, all the data were normalized by subtracting their mean and then dividing by their standard deviation. Varimax rotations were used in this study. Yearly data were organized and analyzed using Statisitca 7.0 (StatSoft, Tulsa, OK, USA). In this study, the critical eigenvalue was set as 0.8. A factor which is only correlated with a single variable dose not contribute to interpreting the data well (Huang *et al.*, 2010). Thus, the number of factors was selected based on these two examination criteria. Although there are several drawbacks for this method (Paatero *et al.*, 2005), this method is fairly easy and widely used. Recently, the Positive Matrix Function (PMF) has been widely applied (Paatero and Tapper, 1994; Zhao *et al.*, 2004); however, PMF is not feasible for this study because of lack of required uncertainty matrix.

RESULTS

Seasonal and Spatial Variations of Trace Metals

Overall, As and Hg, two important pollutants, concentrations observed during this campaign were lower than those measured in previous studies in Taiwan (Table 1) (Hu *et al.*, 2003; Tsai *et al.*, 2003). For all species, the yearly average concentrations were observed highest at the industrial site and the lowest at the wetland site. Although Fe is considered to be a crust element (Huang *et al.*, 2001; Marenco *et al.*, 2006; Nicolás *et al.*, 2008), the significantly higher Fe at industrial site than that at wetland site still implies high Fe resulted from industrial processes, such as

Table 1. Overall trace metals concentrations measured in the study and previous studies.

Site	PM	As (ng/m^3)	Hg (ng/m^3)	Mn (ng/m^3)	Fe (μg/m^3)	Zn (μg/m^3)	Cr (ng/m^3)	Cu (ng/m^3)	Pb (ng/m^3)
Urban-background[1]	PM_{10}	-	-	21 ± 16	1.5 ± 1.9	1.4 ± 2.3	10 ± 11	71 ± 120	47 ± 130
	$PM_{2.5}$	-	-	15 ± 14	1.1 ± 1.4	2.0 ± 1.8	6.2 ± 3.8	21 ± 19	21 ± 27
Urban[2]	PM_{10}	6.0 ± 2.0	2.6 ± 1.6	-	-	-	-	-	-
	$PM_{2.5}$	3.8 ± 1.6	1.8 ± 1.0	-	-	-	-	-	-
Urban[3]	TSP	4.4–18.0	0.07–13.0	19–120	7.20–14.0	0.2 – 0.3	3.4–14.0	100–280	80–110
Suburban/coastal (BS)[4]	TSP	3.1 ± 0.7	0.06 ± 0.03	36 ± 8	2.5 ± 1.0	0.10 ± 0.03	24 ± 7	90 ± 27	34 ± 13
Urban (CH)[4]	TSP	3.2 ± 0.6	0.05 ± 0.02	55 ± 13	2.7 ± 0.7	0.09 ± 0.02	33 ± 7	71 ± 20	29 ± 13
Residential (MH)[4]	TSP	2.8 ± 0.4	0.05 ± 0.02	60 ± 15	2.4 ± 0.7	0.09 ± 0.02	23 ± 8	79 ± 22	32 ± 14
Industrial (QX)[4]	TSP	3.6 ± 0.5	0.07 ± 0.03	82 ± 18	2.8 ± 0.7	0.11 ± 0.03	32 ± 7	110 ± 52	39 ± 11
Wetland (GM)[4]	TSP	2.8 ± 0.3	0.04 ± 0.03	25 ± 8	1.9 ± 0.6	0.07 ± 0.02	18 ± 6	61 ± 17	25 ± 11

1: Rajšić *et al.*, 2008, Serbia; 2: Tasi *et al.*, 2003, Taiwan; 3: Hu *et al.*, 2003, Taiwan; 4: This study, Taiwan; Part of the data has been published in Fang *et al.* (2012).

the ferrous related processes. Zn is regarded as an indicator of mobile sources (Chen *et al.*, 2007; Garg *et al.*, 2000); therefore, it is relatively consistent at these five sites. However, the highest Zn concentrations were also observed at industrial site, and this suggests the Zn emission in this area might be related to the point sources rather than vehicles, such as Cu smelting (Polissar *et al.*, 2001). The significant higher ambient Cu and Pb concentrations at industrial site than others support this hypothesis (Mann Whitney test, *p-value* < *0.05*). All metals show extremely low concentrations but with high variations at Beishi in summer time; however, the reason remained unclear and was probably because of being downwind of the ocean in the summer time. All metals were lower in summer time than other seasons due to higher wet deposition during summer (Fig. 2). In Taichung, in 2010, precipitation amount in summer was approximately 10 times higher than that in winter, and the days of precipitation in summer was twice of those in winter. (http://www.cwb.gov.tw/V7/climate/dailyPrecipitation/dP.htm). This implies trace metals in the atmosphere in this area could be scavenged significantly in summer compared to winter.

PRINCIPAL COMPONENT ANALYSIS

Table 2 lists factor loadings of PCA results, and the total percent explained at these five sites (ranging from 50 to 65%). Except for Beishi which was unraveled with three different factors, only two factors were resolved at other four sites. In Beishi, the first factor is associated with high Mn and Zn concentrations. Both traffic sources and coal-oil combustion could contribute to to atmospheric Mn and Zn concentrations (Lee and Hieu, 2011); however, if we explore in detail of the factor loading, Cr is an indicator of mobile source which is also correlated to this source but low. Based on CPF result, the source was mainly coming from southeast, where Taichung City is located. Taichung is the third largest city in Taiwan with a population of approximately 2,000,000 and significantly impacted by air pollutants emissions from mobile sources. Therefore, this factor is most likely mobile source. Another factor that is associated with As, Cu, and Pb is identified as smelter plants. A previous study reported a nonferrous metal smelter source related to As and Pb using PMF (Lee *et al.*, 1999). The third one is correlated to Hg and Fe. Since primary/secondary pig iron and steel manufacturing plants have been classified as major Hg emission sources (Pirrone *et al.*, 2010), this factor is considered to be coming from ferrous industries. The CPF results also indicate the ferrous/non-ferrous sources originating from southwest where the major industrial region is located.

At the urban site, Changhua, two factors which can explain around 54% of data were indentified, including smelter (both ferrous and nonferrous) and MSWIs. Lee *et al.* (1999) and Polissar *et al.* (2001) both reported that As is an important indicator for smelter with other metals, such as Zn, Mn, and Pb. A recent study also indentified a similar factor of ferrous related source, including Fe, Mn, and Zn (Han *et al.*, 2005). Therefore, the factor related to As, Fe, Mn, and Zn is classified as smelter. However, CPF indicates that the source was from southeast direction, but there is no significant point source in this direction. Hg and Pb are thought to be related to MSWI, and CPF result indicates the source was from east and southeast of the site. There is a MSWI to the east of the site (Fig. 1). The first factor in MeiHe is related to Mn, Fe, Zn, and Cr; as defined above Mn, Zn, and Cr are from mobile source, and Fe and Cr are related to road dust (Manoli *et al.*, 2002). Since CPF shows this source was of east and southeast origin, in which the intersections of two major highways in Taiwan and an urban area (Changhua) are located, respectively. Hence, this factor is classified as mobile sources. The other factor which is correlated with As, Hg, Cu, and Pb and from east and southwest (based on CPF) is classified as MSWIs. There are two and one MSWIs located to the east and southwest of the site, respectively (Fig. 1).

The first factor in Quanxing is associated with As, Mn, Fe, and Zn which has been classified as smelter in previous discussion and originating from southeast of the site; however, similar as site Changhua, no significant point sources exist in this direction. The second factor is related to Hg, Cr, and Pb and from southeast, which is the direction of the major highway intersection in Changhua. Lee and Hieu (2011) reported that Cr and Pb are originated from

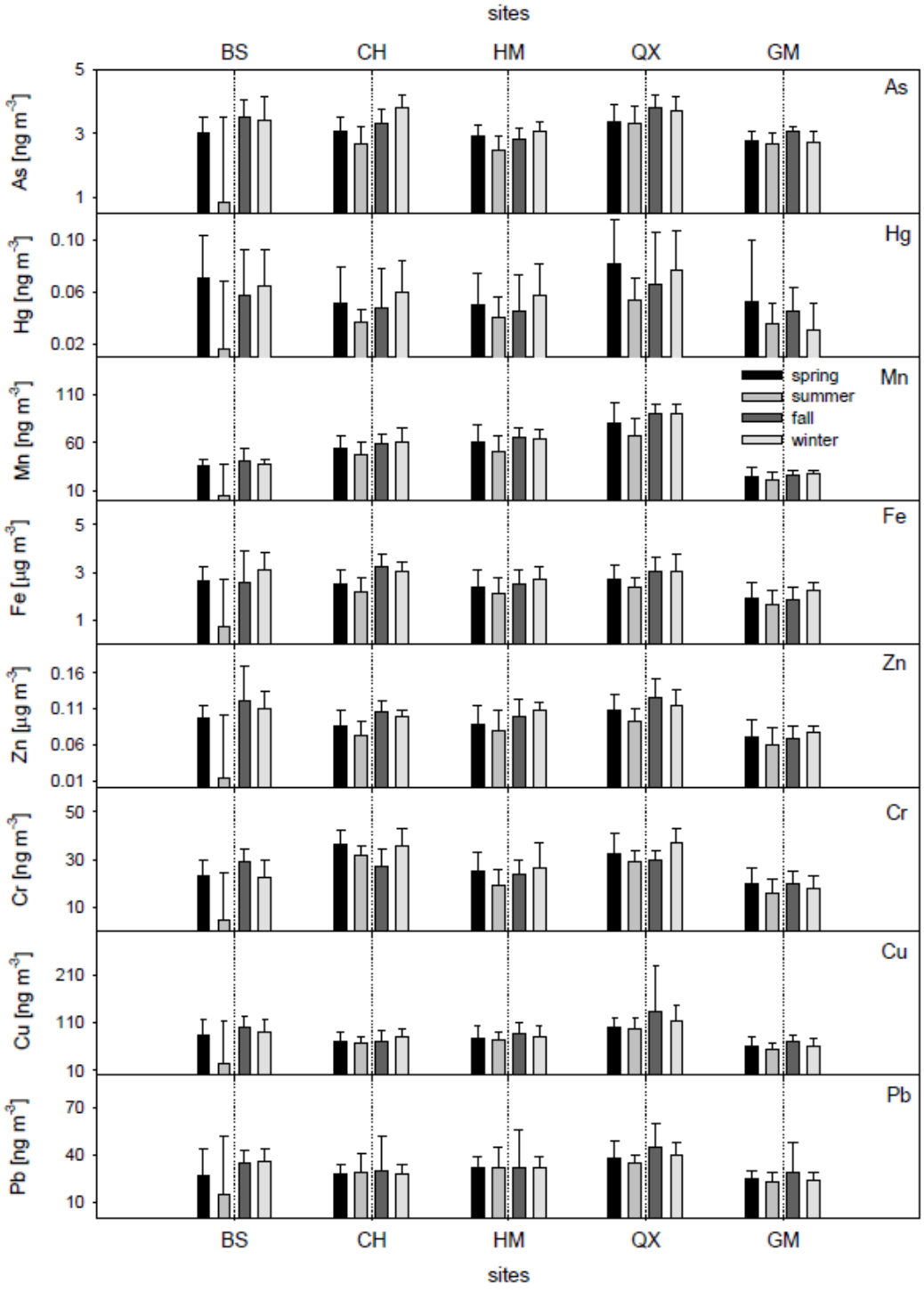

Fig. 2. seasonal patterns of ambient trace metals concentrations at five sites in Taichung. Part of the data has been published in Fang *et al.* (2012).

mobile sources. However, in Taiwan the gasoline containing lead has been banned since 2000; therefore, most likely Pb does not represent mobile source. Additionally, Hg was never discovered to be significantly correlated to mobile sources (typical correlation coefficient of 0.2–0.3 (Huang *et al.*, 2010)). Therefore, this factor might not be mobile source or involve two different sources. In Geomei, the first factor is identified as mobile source with Mn, Fe, Zn, and Cr and points to Taichung City. The second factor is MSWIs and most likely from those facilities located in Taichung city.

DISCUSSIONS

Trace metals concentrations measured at five different sites in central Taiwan show clear seasonal patterns of

Table 2. PCA factor loading from the eight different trace metals measured in this study. EV and PT represent as the explained variance and the total percent explained.

Factor	BS			CH		HM		QZ		GM	
	1	2	3	1	2	1	2	1	2	1	2
As	0.42	**0.56**	–0.01	**0.66**	0.32	0.12	**0.61**	**0.50**	0.03	0.11	**0.69**
Hg	–0.03	–0.09	**0.86**	0.34	**0.51**	0.19	**0.60**	0.24	**0.57**	0.28	0.01
Mn	**0.84**	0.01	0.25	**0.76**	–0.02	**0.87**	0.12	**0.87**	0.15	**0.97**	0.05
Fe	0.19	0.25	**0.54**	**0.84**	–0.05	**0.89**	–0.11	**0.84**	–0.01	**0.90**	–0.03
Zn	**0.84**	–0.06	–0.23	**0.87**	0.04	**0.86**	0.09	**0.87**	0.12	**0.97**	0.00
Cr	0.46	–0.12	0.29	0.28	0.08	**0.76**	–0.06	0.00	**0.77**	**0.88**	0.02
Cu	–0.02	**0.76**	–0.02	0.11	**0.73**	–0.04	**0.58**	0.12	0.28	0.05	**0.76**
Pb	–0.17	**0.76**	–0.05	–0.09	**0.84**	–0.10	**0.78**	0.21	**0.64**	–0.10	**0.72**
EV	1.86	1.56	1.25	2.68	1.61	2.93	1.71	2.58	1.45	3.57	1.59
PT	0.23	0.20	0.16	0.34	0.20	0.37	0.21	0.32	0.18	0.45	0.20

lower concentrations in summer than other seasons. An obvious spatial distribution was observed for all trace metals with highest values in industrial area and lowest found at a back ground site, wetland environment.

PCA results indicate that mobile source, MSWIs, and smelters are the major trace metals emission sources in central Taiwan, and some of these conclusions are also supported by the CPF results which were used to identify source locations. However, one factor remains unclear factor loading with its corresponding CPF plot. A potential smelting source from southeast cannot be linked to any point source based on the current industrial inventory, and this suggests the factor might not be listed in the inventory or from another industrial area. There are two major sources in this area not accounted for, including a CFPP and a steeling plant. Both of them are located to the west/north of four of the sites, except Gaomei. In central Taiwan, the prevailing wind direction was from southwest; it might be the reason why these two sources did not stand out from PCA and CFP analyses.

CONCLUSIONS

Eight different trace metals were measured from TSP samples collected using PS-1 at five different sites in central Taiwan. The results suggest that trace metals concentrations in this area are influenced by mobile sources, MSWIs, and smelters. However, two important point sources in that region did not reflect their contributions in particular to those selected sampling sites in this study. Taiwan is an intensively industrialized country, especially in central Taiwan. There are several concentrated industrial areas in central Taiwan with the intensive mobile emissions and heavy-duty point sources, such as CFPP and steeling plants. Furthermore, this particular region not only consists of urban areas, agricultural areas (potential agricultural waste burning sources), but also is close to the ocean. The unique combination in topography and meteorology along with atmospheric chemistry lead to even more complicated air pollutant transformation and transport after being emitted. All these make the sources identifications and receptor modeling results difficult to resolve and explain, especially in Taiwan, where national emission inventory is not documented. Further investigations with short sampling interval and effort are merited in order to improve our understanding of adverse health effects from exposure to particulate trace metals.

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