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Chemical Compositions of $PM_{2.5}$ in Residential Homes of Southern Taiwan

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Abstract

Aerosol samples of fine particles ($PM_{2.5}$) were collected simultaneously from inside and outside 16 residential homes in Tainan City of southern Taiwan. The collected samples were analyzed for the carbon content and 16 elements including Be, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, Hg and Pb. The concentrations of mean elemental carbon (EC) and organic carbon (OC) of the indoor and corresponding outdoor $PM_{2.5}$ were approximately the same and within the range of 10–12 $\mu g/m^3$. The indoor/outdoor (I/O) ratios of EC and OC varied at 0.35–3.86 and 0.27–2.44, respectively. Metals such as Ca, Fe, Pb, and Zn were most abundant among the elements examined, while elements such as Be, Co, Hg, Cd were found in the smallest quantities. Using Fe as the reference element, the enrichment factors (EFs) varied from less than unity to more than 60,000. According to the degree of enrichment, As, Se, Cd, Sb, Hg were found to be the most enriched in $PM_{2.5}$. The results suggest that the anthropogenic combustion sources such as coal combustion, municipal waste incineration, vehicular emission as well as building construction/renovation are the major sources of indoor $PM_{2.5}$ in Tainan City.

Keywords: Chemical composition; $PM_{2.5}$; Indoor air quality; I/O ratio.

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INTRODUCTION

Indoor air quality has become a critical issue and a public health concern in the modern society. Increasing public awareness is focusing on this issue as more and more individuals spent time inside than outside (Robison and Nelson, 1995), particularly those who are most susceptible to the effects of poor air quality, such as the elderly, the young, and those with poor health. Moreover, air pollutants are frequently accumulated in much higher concentration indoors than outdoors (Lee *et al.*, 2002). Airborne pollutants originating from indoor sources such as smoking, cooking and cleaning predominately contribute to poor indoor air quality (Wallace, 1996). Besides, indoor air quality can be affected by pollutants emitted from outdoor sources like vehicles and industries through infiltration. Additionally, indoor air quality can also be affected by the scavenging mechanism of air pollutants and the ventilation condition in the indoor environment.

Chemical compositions of inhaled particles play an important role in the causation of adverse health effects. For example, organic carbon (OC) may contain polycyclic aromatic hydrocarbons and other components with possible mutagenic and carcinogenic effects (Ho *et al.*, 2004). Elemental carbon (EC) formed from incomplete combustion of carbon-based fuels has been found to be associated with significantly increased risk of death from lung cancer and other severe respiratory diseases (Frazer, 2002). Particulate matter may contain some elements which are highly toxic or even carcinogenic. For example, elements such as antimony (Sb), arsenic (As), beryllium (Be), cadmium (Cd), chromium (Cr), Cobalt (Co), lead (Pb), manganese (Mn), mercury (Hg), nickel (Ni) and selenium (Se) have been deemed hazardous air pollutants (HAPs) under the U.S. Clean Air Act Amendments of 1990. Furthermore, As, Be, Cd, Cr, Co and Ni have been classified by the International Agency for Research on Cancer (IARC) as group 1 human carcinogens or group 2B possible human carcinogens (IARC, 1980).

Numerous epidemiological studies have investigated the airborne particulate matter (PM) on human health (Pope *et al.*, 1999), several of which highlight the health significance of fine particles smaller than 2.5 μm in aerodynamic diameter as they can penetrate deeply into the respiratory tract (McDonnell *et al.*, 2000; Schwartz and Neas, 2000; Andersen *et al.*, 2001). The USEPA conducted a human exposure and epidemiological study to investigate $PM_{2.5}$ exposure and health effects (Williams *et al.*, 2000a-c). A number of researches have been focused on the mass concentrations and the carbonaceous species for indoor air quality pertaining to $PM_{2.5}$ (Neas *et al.*, 1994; Funasaka *et al.*, 2000; Jones *et al.*, 2000; Gotschi *et al.*, 2002; Li and Lin, 2003; Ho *et al.*, 2004; Na and Cocker, 2005). Several studies have characterized indoor $PM_{2.5}$ on chemical speciation (Hirosihi *et al.*, 1994; Yakovleva *et al.*, 1999; Chao and Wong, 2002; Graney *et al.*, 2004). Only limited investigations have been performed to evaluate both carbon and chemical profiles of indoor $PM_{2.5}$ (Geller *et al.*, 2002).

With the emphasis on indoor $PM_{2.5}$, the objectives of this study are to provide quantitative information on the concentrations of organic and elemental carbon as well as elements and to investigate the indoor/outdoor relationship of chemical compositions in residential homes of Tainan in southern Taiwan. Tainan is home to over 700,000 residents and a number of family-based factories are located in the city. Some of them are traditional metal industries such as electroplating and welding. Few of these family-based factories are equipped with air purifying devices because of their small industrial scale. Additionally, several large industrial parks, such as the Ann-Ping industrial area and Yung-Kung industrial area, are near the downtown area. There is also a large power plant in the vicinity of the city and biomass burning in the open field can be observed occasionally. In this study, we simultaneously measured indoor and outdoor air of 16 residential homes for EC and OC as well as 16 elements including Be, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, Hg and Pb.

METHODS

This field study was conducted in Tainan during April and June 2001. Tainan City encompasses seven administration districts including Ann-Nan Region, Ann-Ping Region, West Region, North Region, South Region, Middle Region and East Region. Sixteen residential homes with natural ventilation were selected from five densely populated districts for air sampling (Fig. 1). The population in the area investigated accounts for over 70% of the total population in Tainan.

The personal environmental monitoring (PEM) impactor (MSP, Minneapolis, MN, USA) was utilized for gathering $PM_{2.5}$ aerosols at a flow rate of 10 L/min. The PEM impactor is a single stage impactor with ten circular jets that provide a sharp cut size at 2.5 μm. The impaction surface is an oil-soaked, porous stainless steel plate. For each sample, the impaction surface was given a light film of mineral oil to minimize particle bounce. Particles > 2.5 μm were collected on the impaction surface, whereas those < 2.5 μm were collected in a quartz-fiber filter (2500 QAT-UP, Pallflex Products Corp., Putnam, Connecticut, U.S.A.) and a Teflon filter (Zefluor, 37 mm diameter, 1 μm pore size, Gelman Sciences, Ann Arbor, Michigan, U.S.A.) via filtration. Before and after the field sampling, the filters were placed in a room of temperature 25℃and relative humidity of 50%. Duplicate air samples were simultaneously obtained in the living room and a corresponding outdoor site such as on a balcony or in front door area for each house. Samples were collected at the height of approximately 1.2 m above the ground level to simulate the breathing zone (Li and Lin, 2003; Ho *et al.*, 2004) for 24 h for each sample.

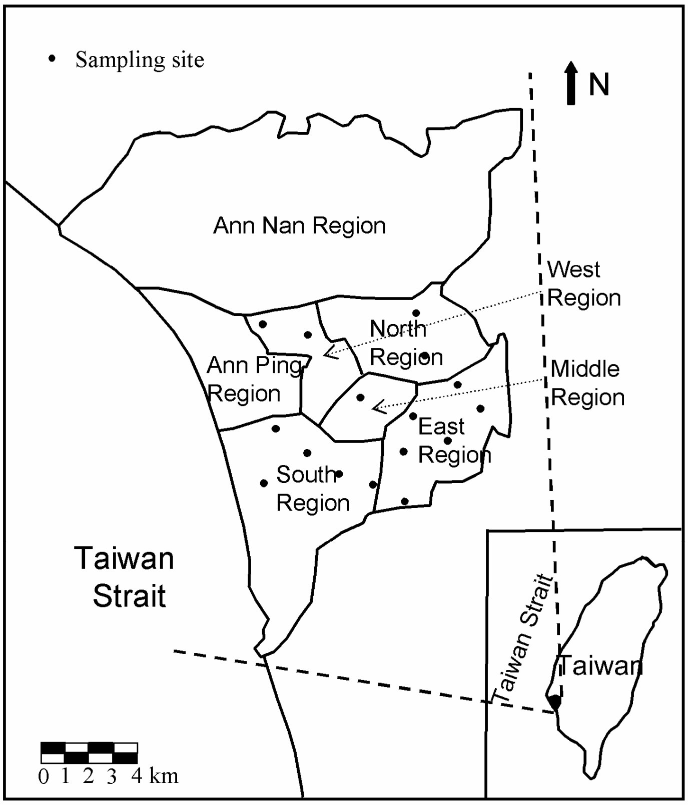

Fig. 1. Map of sampling site in Tainan City.

Particle samples collected on quartz fiber filters were examined for total carbon (TC) and elemental carbon content using a Total Carbon Analyzer coupled with a Solid Sample Module (SSM) (Shimadzu, Japan). The thermal/optical protocol for determining particle carbon content is similar to those described by other researches (Cadle *et al.*, 1983; lavanchy *et al.*, 1999; Li and Lin, 2003). To reduce the background level of carbonaceous species in the quartz filter, filters were preheated at 900℃ for 4 h prior to sampling. After sampling, each filter was weighed to determine the particle mass concentration and then cut into two halves. One half of the filter was placed in a sample boat and burned at 950℃ in the combustion chamber of the SSM to determine the mass of TC. The combustion products, water vapor and carbon dioxide, were led to a drain separator that trapped the water. Carbon dioxide was transferred into the analyzer to determine the concentration of CO_2 by non-dispersive infrared radiation (NDIR) according to the Lambert-Beer law. The mass of TC in each particle sample was calculated using a calibration curve using glucose as the standard solutions. The other half of the filter was conditioned at 350℃ for 45 minutes with helium as the carrier gas to purge organic carbon, and then placed in a sample boat and analyzed for remaining carbon content (EC) using the same procedure as that for TC. The mass of OC was the difference between the mass of TC and EC.

Analysis of the elements was conducted at the analytical laboratory of Joint Center for High Valued Instruments at National Sun Yat-Sen University, Kaohsiung, Taiwan by an inductively coupled plasma-mass spectrometer (ICP-MS, SCIEX-Elan-5000, Perkin-Elmer, Toronto, Canada). Particle samples collected on Teflon filters were prepared using a microwave-digestion system (CEN MDS-2000, Milestone, USA) for subsequent spectrometric analysis. Maximum system output power of the system is 630 W. A rotating turntable capable of holding 12 closed digestion vessels was used to ensure even sample heating. The Teflon filters with particulate samples were folded and placed into the closed 100-ml PTFE-lined digestion vessels with 4.5 ml HNO_3-HF (8:1, v/v) and digested in the microwave oven. The digestion process consisted: first with 70% power for 10 minutes under 40 psi air pressure, then at 90% power for 50 minutes under 160 psi. After digestion and cooling, the acid solution of the sample was directly diluted to 25 ml with distilled water and kept in an acid-washed polyethylene bottle. The acid solutions of the samples were then subjected to analysis with an ICP/MS for the elements including Be, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, Hg and Pb. The detection limits for most elements analyzed here by ICP-MS varies at 0.03-0.1 ng/ml in the solutions, except that for Ca (10 ng/ml) (Wang *et al.*, 1995; Jalkanen and Hasanen, 1996). Assuming a 14.4 m^3 sample volume, he equivalent concentrations in air are 0.052-0.17 ng/m^3 for digested solution diluted to 25 ml in this study. The detailed analytical procedures of elements in Teflon filters and QA/QC can be found in the work prepared by U.S. Environmental Protection Agency (USEPA, 2005).

RESULTS AND DISCUSSION

For $PM_{2.5}$ samples from the 16 selected residential homes, the averaged concentrations of OC indoors and outdoors were 12.1 ± 5.6 μg/m^3 and 9.8 ± 4.0 μg/m^3 while mean concentrations of EC indoors and outdoors were 11.1 ± 4.5 μg/m^3 and 10.9 ± 4.5 μg/m^3, respectively. Comparative data of carbon species for indoor and outdoor $PM_{2.5}$ in different areas are summarized in Table 1. Thermal analysis was used for carbon analysis for the studies in Table 1. To split the OC and EC, different thermal protocol was adopted by researchers and was described in details by Chow *et al.* (2005). The thermal protocol used in this study was similar to that used in the study of Li and Lin (2003). During the thermal process of OC/EC split, some OC were pyrolitically converted to EC. The EC data reported by Ho *et al.* (2004) and Na *et al.* (2005) have been corrected for the pyrolyzed OC. Therefore, the EC data reported by Li and Lin (2003) and this study have to be considered with conservation. However, it was found that both indoor and outdoor EC concentrations measured in Tainan were two times higher than those measured in Taipei (Li and Lin, 2003). Elemental carbon can be generated by the combustion of fossil fuel from various sources such as coal-burning industry and diesel-powered vehicles (Gotschi *et al.*, 2002;

Funasaka *et al.*, 2000). Accordingly, the measured EC may be attributed to local combustion sources such as diesel automobiles and the power plant located south-west of Tainan City.

Table 1. Mean organic carbon (OC) and elemental carbon (EC) concentrations of $PM_{2.5}$ samples in μg/m^3 at various indoor and outdoor sites.

	Indoor			Outdoor			OC	EC
	OC	EC	OC/EC	OC	EC	OC/EC	I/O	I/O
This study	12.1	11.1	1.2	9.8	10.9	1.0	1.3	1.2
Li and Lin, 2003	12.3	5.1	2.9	10.5	4.8	2.0	1.2	1.1
Ho et al., 2004	11.3	4.8	2.7	12.6	6.4	2.0	1.0	0.8
Na et al., 2005	14.8	2.0	7.4	12.3	2.5	5.0	1.4	0.8

Due to the high EC levels in this study, the indoor and outdoor OC/EC ratios were the lowest as listed in Table 1 and the results of indoor and outdoor OC/EC ratios were 1.2 and 1.0, respectively. However, the mean outdoor OC and EC concentrations of $PM_{2.5}$ measured in this study were comparable to those measured in urban air by Yuan *et al.* (2002) in Kaohsiung City, which is 30 kilometers away in the south of Tainan. Yuan *et al.* reported that the EC and OC concentrations of ambient $PM_{2.5}$ were 10.7 ± 3.6 μg/m^3 and 11.9 ± 3.7 μg/m^3, respectively.

Regarding the indoor/outdoor relationship of carbon species, the I/O ratios of $PM_{2.5}$ for OC and EC varied at 0.3-2.4 with a mean value of 1.3 and 0.4-3.9 with a mean value of 1.2, respectively. Chuang *et al.* (1991) suggested that the I/O ratio above 3 can be considered as an indicator of the existence of the indoor sources. Our results showed that the averaged I/O ratios for OC and EC were slightly larger than 1, indicating that there are no significant indoor sources. The I/O results were also comparable to the results obtained in residential homes of northern Taiwan (Li and Lin, 2003).

Paired t-test indicated that there is no significant difference between concentrations for both OC and EC indoors and outdoors. Fig. 2 shows a plot of indoor-to-outdoor OC and EC concentrations. As for OC, it was found that a weak correlation between indoor and outdoor levels with the coefficient of determination (R^2) of 0.15. Moreover, no significant linear correlation existed between indoor and outdoor EC levels.

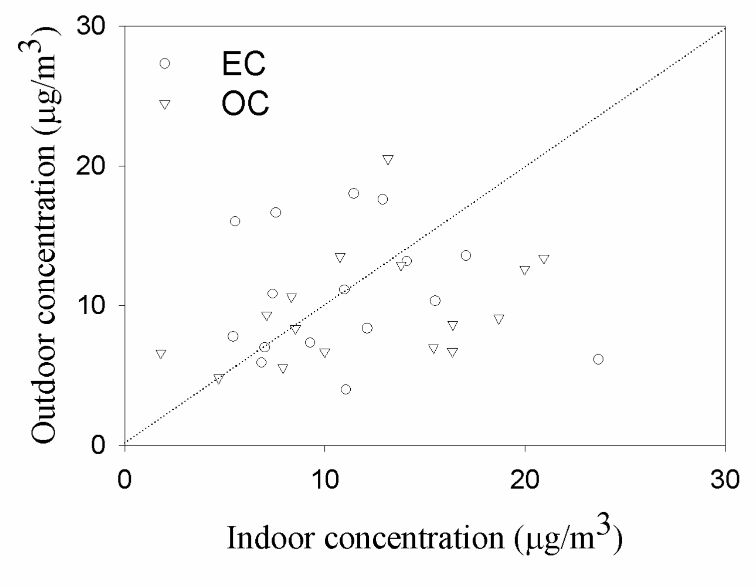

Fig. 2. Indoor versus outdoor carbonaceous concentrations in $PM_{2.5}$ aerosols.

The indoor and outdoor elemental data for $PM_{2.5}$ samples are shown in Table 2. The field blanks of the filters were examined for trace element impurities. The averaged values of field blank data were used to correct the final analytical results. Generally, the elemental composition of $PM_{2.5}$ measured in this study consists with a typical abundance order found in ambient air (Fernádez *et al.*, 2001). Elements Ca, Fe, Zn and Pb were most abundant, whereas elements Be, Co, Hg, Cd were least abundant. The soil-related elements, Ca and Fe, were typically found in coarse particles and the results of this study demonstrated that these two elements were also predominant in fine particles. The indoor and the corresponding outdoor concentrations were quite comparable and were in the same order of magnitude. For most of the elements evaluated in this study, no significant difference was observed between the indoor and outdoor concentrations by paired t-test.

Both indoor and corresponding outdoor elemental concentrations in $PM_{2.5}$ measured in this study were generally higher than those reported in other areas (Yakovleva *et al.*, 1999; Chao and Wong, 2002). Table 3 lists the comparative data on average elemental concentrations of indoor and outdoor $PM_{2.5}$ in ng/m^3. The measured concentrations were about in the same order of magnitude for the studied elements (Ca, V, Cr, Mn, Fe, Ni, Cu, Zn and Pb) except for Ca. Calcium was the most abundant element among measured elemental species. Mean Ca concentrations of indoor and the corresponding outdoor $PM_{2.5}$ were approximately 7.6 and 6.0 $\mu g/m^3$ and were much higher than those reported in other areas (Hiroshi *et al.*, 1994; Yakovleva *et al.*, 1999; Hidy *et al.*, 2000; Chao and Wong, 2002; Geller *et al.*, 2002). Kasahara (1992) reported that airborne Ca levels may be attributed to emissions of cement dust from building construction/renovation. Moreover, the corrosion of the cement is a common phenomenon in the residential houses of Tainan and therefore may contribute to the elevated concentration of Ca in the air.

Table 2. Indoor and outdoor elemental concentrations of $PM_{2.5}$ samples in ng/m^3.

	Indoor				Outdoor			
Element	Range	Median	Mean	S.D.	Range	Median	Mean	S.D.
Be	0.1-2.0	0.9	1.0	0.8	0.1-1.5	0.6	0.7	0.6
Ca	960.9-22130.2	6159.9	7666.0	7718.2	686.3-29015.1	1810.2	5970.0	8547.7
V	1.9-13.9	6.3	7.0	3.6	1.5-14.0	6.2	6.9	3.7
Cr	0.9-41.0	13.2	15.7	11.8	4.1-46.2	10.0	18.2	15.2
Mn	2.9-53.5	17.5	20.6	17.2	1.0-65.4	16.5	20.0	18.1
Fe	79.0-1531.1	362.6	560.9	486.2	21.1-1812.5	327.4	552.3	516.7
Co	0.1-2.2	0.4	0.7	0.7	0.1-4.0	0.6	1.0	1.0
Ni	0.1-12.7	4.8	5.5	5.0	0.7-15.5	2.3	5.4	5.6
Cu	5.8-111.4	24.0	34.5	25.9	4.4-215.2	41.9	49.8	49.3
Zn	9.8-265.3	89.1	106.4	67.5	1.2-212.2	110.8	116.7	65.3
As	2.5-110.2	18.2	26.1	31.5	0.1-143.5	12.5	40.5	54.3
Se	1.1-14.5	4.8	5.6	3.4	1.4-12.9	4.2	4.6	2.9
Cd	0.4-4.9	2.2	1.9	1.4	0.4-4.5	1.6	1.9	1.3
Sb	1.7-31.4	7.4	10.3	8.9	1.5-70.4	12.0	18.2	21.2
Hg	0.5-4.1	1.6	1.9	1.2	0.1-12.0	1.9	2.3	2.7
Pb	13.0-295.7	52.9	97.5	89.0	5.6-656.1	57.6	136.8	189.1

For the relationships between the elements, Table 4 lists the correlation coefficients that are > 0.8 for indoor and outdoor $PM_{2.5}$ samples. Elements Be and Ca were strongly correlated with R value > 0.9 for both indoor and outdoor samples. This is attributed to the fact that both Be and Ca occur naturally as components of rocks. Elements Fe and Mn typically originate from the same sources such as soil and the steel industry and were strongly correlated as expected. Furthermore, Pb, As, and Sb were strongly correlated for outdoor $PM_{2.5}$ samples, indicating that emissions from the combustion process of fossil fuel.

Enrichment factors (EFs) for each element were calculated to identify potential particle sources. Based on the average elemental concentrations, the EFs were calculated by dividing the ratio of the average elemental concentration of target species to reference species in the aerosol sample by the ratio of the target species to reference species in the crust. Gao *et al.* (1992) suggested that Al, Fe, Se and Ba were commonly considered as the reference elements of crustal material. Using Fe as the reference element, Fig. 3 presents EFs for the analyzed elemental species of indoor and outdoor $PM_{2.5}$ samples relative to the elements of average crustal rock (Mason, 1966). The measured EFs varied widely from less than unity to more than 60,000. Elements with an EF

Table 3. Comparative data on average elemental concentrations of indoor and outdoor $PM_{2.5}$ in ng/m^3.

	Indoor			Outdoor		
Element	a	b	c	a	b	c
Ca	7666.0	308.4	138.2	5970.0	251.6	172.3
V	7.0	4.9	4.6	6.9	5.6	4.9
Cr	15.7	5.0	NA	18.2	2.9	NA
Mn	20.6	8.9	10.7	20.0	11.4	16.7
Fe	560.9	280.2	140.1	552.3	343.7	211.6
Ni	5.5	2.7	1.8	5.4	2.7	2.1
Cu	34.5	10.6	10.7	49.8	9.1	13.0
Zn	106.4	38.8	130.6	116.7	40.2	144.9
Pb	97.5	18.0	88.1	136.8	21.6	103.6

a This study

b Yokovleva et al., 1999

c Chao et al., 2002

Table 4. Correlation coefficients (R) of ≥ 0.8* between analyzed elemental species in indoor and outdoor $PM_{2.5}$ samples.

	Indoor			Outdoor	
Element	R ≥ 0.9	0.9 > R ≥ 0.8	Element	R ≥ 0.9	0.9 > R ≥ 0.8
Be	Ca		Be	Ca	As
V		Mn, Fe, Co	V		Mn, Fe
Cr	Pb	Mn, Fe	Cr		Fe, As, Sb, Pb
Mn		Fe, Pb	Mn	Fe	As, Sb, Pb
Co		Zn	Fe		As, Sb, Pb
Cu		Se	As	Sb, Pb	
			Sb	Pb	

greater than unity are assumed to originate from sources other than crustal dust. However, EFs are typically not assumed to be resolved within an order of magnitude (EF < 10) from crustal materials (Fernádez *et al.*, 2001). According to the degree of enrichment, the studied elements were grouped as follows:

Highly enriched (EF ≥ 1000): As, Se, Cd, Sb, Hg

Moderately enriched (EF 100- < 1000): Zn, Pb

Slightly enriched (EF 10- < 100): Be, Ca, Cr, Cu
Minimally enriched (EF < 10): V, Mn, Co, Ni

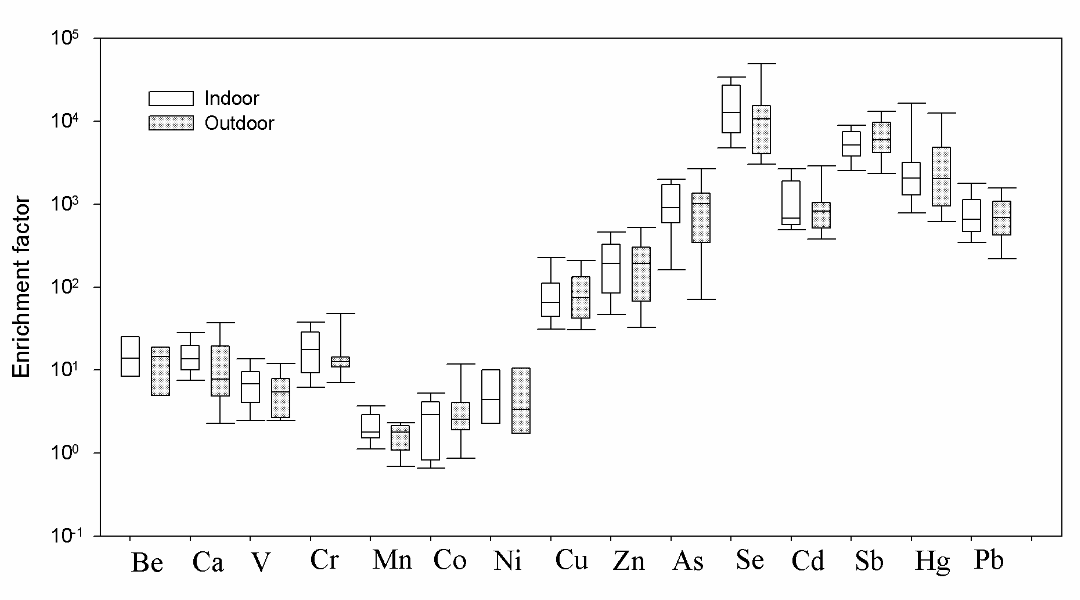

Fig. 3. Enrichment factors of analyzed elemental species in indoor and outdoor $PM_{2.5}$ samples (Fe as the reference element). Boxes represent the 25th, 50th, and 75th percentiles and lines extend to 5th and 95th percentiles.

Highly enriched elements such as As, Se, Cd, Sb, Hg in this study were generally volatile elements derived from high-temperature processes (fossile fuel combustion and smelting) (Berg *et al.*, 1994). Both Cd and Hg can result from emissions of industrial sources such as metal smelting processes, whereas As, Se and Sb can be emitted into the air during coal burning. High enrichment of As, Se, and Sb may be attributed to emissions from the coal-fired power plant located south-west of Tainan City and brought to the city by prevailing winds during the sampling season.

The element Se is the most strongly enriched element in coal. Selenium compounds are released into the air during burning of coal and petroleum fuels, and during smeltering and refining of other metals. Elevated levels of Se exposure can cause reproductive failure; however, there is no data available for a threshold value of airborne Se. Moreover, the bioconcentration factor of Se is fairly high—approximately 50,000 for marine fish (http://www.epa.gov/ogwdw000/dwh/t-ioc/selenium.html). The fish-culturing industry along the coast near Tainan City is a major industry. Airborne Se can fall into the ocean via dry deposition and/or wet deposition, implying that fish may accumulate a hazardous dose of Se and pose a health risk through food chain.

In this study, outdoor Sb and Pb were strongly correlated as shown in Table 4. Notably, Sb, Zn, and Pb were considered elements indicative of refuse combustion (Kasahara, 1992). A recent study investigating heavy metals emitted from small incinerators identified the enrichment of Cd, Cu, Pb, and Zn in $PM_{2.5}$ among test metals including Cu, Cd, Mn, Cr, Mg, Pb, Zn (Yoo *et al.*, 2002). Hence, enriched Sb, Cd, Pb and Zn indicated that municipal waste incineration may contribute to atmospheric fine particles in the urban air of Tainan city. Moreover, Huang *et al.* (1994) indicated that emissions from motor vehicles are the potential sources for atmospheric Sb and Zn in the urban areas and Prati *et al.* (2000) suggested that Zn, Pb and Cu usually originate from traffic sources. Therefore, enriched Sb, Zn, Pb, and Cu are likely due to emissions of fine particles from traffic sources.

CONCLUSIONS

Carbonaceous and elemental concentrations of indoor and outdoor $PM_{2.5}$ aerosols were characterized in 16 residential homes in Tainan City of southern Taiwan. The EC and OC for both indoor and outdoor $PM_{2.5}$ samples were approximately at the same level and within the ranges of 10-12 $\mu g/m^3$. The I/O ratios for EC and OC were 0.35-3.86 and 0.27-2.44, respectively. The elemental composition of $PM_{2.5}$ follows a common abundant order. For the elements examined, Ca was the most abundant and at the $\mu g/m^3$ level while other elements were at the ng/m^3 level or less. In addition to Ca, elements such as Fe, Pb, and Zn were also abundant, whereas elements such as Be, Co, Hg and Cd were in the smallest quantities. Using Fe as a reference element, the EFs varied from less than unity to more than 60,000. According to the enrichment degree, the evaluated elements were grouped as follows: highly enriched ($EF \geq 1000$), As, Se, Cd, Sb, and Hg; moderately enriched (EF 100– < 1000), Zn and Pb; slightly enriched (EF 10– < 100), Be, Ca, Cr and Cu; and minimally enriched ($EF < 10$), V, Mn, Co and Ni. The results suggest that the anthropogenic combustion sources such as coal combustion, municipal waste incineration, vehicular emission as well as building construction/renovation are the major sources of $PM_{2.5}$ in Tainan City.

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