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Characteristics of Respirable Particulate Metals Emitted by a Beehive Firework Display in YanShuei Area of Southern Taiwan

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ABSTRACT

This study investigates metals in the $PM_{1.0}$ and $PM_{2.5}$ collected using a micro-orifice uniform deposition impactor (MOUDI) sampler in the YanShuei area of southern Taiwan during a beehive firework display. The results of sample analyses indicate that during the beehive firework display, the ratios of metal concentrations in $PM_{2.5}$ (D) to the background level (B) at leeward sampling site were 1,828 for Ba, 702 for K, 534 for Sr, 473 for Cu, 104 for Mg, 121 for Al, and 98 for Pb. The corresponding data for $PM_{1.0}$ were 3036, 838, 550, 676, 594, 190, and 126, respectively. According to the results of metal composition ratio, Principal Component Analysis (PCA), and upper continental crust (UCC) analyses, the concentrations of particle-bound Al, Ba, Cu, K, Mg, Pb, and Sr increased during the beehive firework displays, suggesting that firework-display aerosols contained abundant metal elements of Al, Ba, Cu, K, Mg, Pb, and Sr. Before (background), trial, during, and after the beehive firework display, the Ba, K, Cu, Mg, Pb, and Sr (commonly regarded as firework display indicator elements) accounted for 0.520, 2.45, 26.4 and 0.849% mass of PM_1, respectively, while for $PM_{2.5}$ the corresponding data were 0.777, 2.32, 23.8, and 0.776%, respectively.

Keywords: Beehive fireworks display; PM_1; $PM_{2.5}$; Metals; Short-term pollution.

INTRODUCTION

The short-term effects of air pollution on health have attracted increasing attention in recent years. The extensive use of pyrotechnics in large celebratory events frequently degrades short-term air quality significantly, possibly harming human health (causing chronic lung diseases, cancer, neurological and haematological diseases, for example) (Smith and Dinh, 1975; Clark, 1997; Godri *et al.*, 2010; Moreno *et al.*, 2010; Caballero, *et al.*, 2015; Robles, *et al.*, 2015). Fire work displays are known to increase ambient fine particle concentrations and fine-particulate metals (Vecchi *et al.*, 2008; Lancaster *et al.*, 1998; Perry, 1999; Kumara *et al.*, 2016). The complex nature of the particles that are emitted during fireworks may have adverse health, effects as reported by Ravindra *et al.* (2001). Furthermore, in the 2007 Montréal International Fireworks Competition, $PM_{2.5}$ levels of 10,000 μg m^{-3} were reached, equal to approximately roughly 1,000 times the background level (Alexandre *et al.*, 2010). Zhang *et al.* (2010) reported the measurement of the number concentrations and size distributions of aerosol particles with aerodynamic diameters in the range of 10 nm to 10 μm during the Chinese New Year's firework event in Shanghai, China. Particle concentrations during the peak hour of firework celebrations were approximately three times higher than on the preceding day, with a clear shift in the particles from nuclei mode (10–20 nm) and Aitken mode (20–100 nm) to accumulation mode (0.5–1.0 μm).

More than 10 million firecracker/firework rockets displayed in the Lantern festival night every year in the YanShuei area. Therefore, the firecracker/firework display emitted abundant $PM_{2.5}$ at the short-term. However, the mass concentrations and chemical compositions (metal components/concentrations) of $PM_{1.0}$, $PM_{2.5}$, and PM_{10} from beehive firework displays have seldom been investigated. Accordingly, this study investigates the mass concentrations in $PM_{1.0}$, $PM_{2.5}$, and PM_{10}, as well as the metal components (Al, Ba, Ca, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sr, and Zn) and concentrations in particles that were collected in the YanShuei area of southern Taiwan. The size distributions and cumulative mass fractions of metals in particles in each size range are

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obtained from the samples that were collected using a MOUDI sampler. The results of an analysis show that a beehive firework display emits significant amounts of PM_1 and $PM_{2.5}$ degrading, short-term air quality, requiring that related health concerns be addressed.

MATERIALS AND METHODS

Collection of Particulates

Atmospheric particulate samples were collected in the YanShuei area of southern Taiwan during the Lantern Festival from February 21 to 25 in 2013. The windward sampling site was located on the roof of a three-story building (9 m height) in the Wumiao Temple, roughly 50 m north to the major beehive fireworks display site, while the leeward sampling site was located on the roof of a four-story building (12 m height) in the YanShuei police station, roughly 300 m south to the major beehive fireworks display site. The YanShuei beehive fireworks display events occurred within the four stages of our experimental periods. In this investigation, it is regarded that February 21st–22nd, 23th–24th, 24th (18:00–24:00), and 25th 2013 were the before (background), trial, during, and after beehive fireworks display periods, respectively. The mean air temperature, relative humidity, and wind speed were 19.1°C (14.4–26.4°C), 78.9% (56.0–94.0%), and 0.54 (0.0–2.2) m s^{-1}, respectively, during the sampling period (without any rain).

A MOUDI (Model No.100; MSP Co., Minneapolis, MN) sampler equipped with Teflon filters (with diameters of 37 mm) was used to collect size-resolved aerosol samples. These impactors effectively separated the particulate matter into 10 ranges (at 50% efficiency) with the following equivalent cut-off aerodynamic diameters; 18–10, 10–5.6, 5.6–3.2, 3.2–1.8, 1.8–1.0, 1.0–0.56, 0.56–0.32, 0.32–0.18, 0.18–0.1, and 0.1–0.056 µm. Accordingly, the particles were divided into three size groups - PM_{10}, fine ($PM_{2.5}$), and accumulation ($PM_{1.0}$) particles. The sampling flow rate for the MOUDI was 30 L min^{-1}.

Silicon grease was applied to the surface of each filter installed in the MOUDI sampler, and the greased filter-strips were baked in a 60°C oven for 90 min to stabilize the silicon grease before sampling to minimize particle bounce between the different stages of the MOUDI during the sampling. Before and after each sampling, the filters were dried for 24 h in a desiccator at 25°C in 40% relative humidity. They were then weighed on an electronic balance (AND HM202) with a resolution of 10 µg. The suspended particulate matter concentration was determined by dividing the particle mass by the volume of sampled air.

Metals Analysis and Quality Control

Metals Analysis by Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES) Particles were digested in a 1,600 W microwave oven (Mars, microwave digestion system, CEM) according to Yang and Swami (2007) and Tsai et al. (2003) to ensure accurate and reliable analysis of metals in the particles. The digested solution was a mixture of 10.0 mL (65% HNO_3 and 37% HCl) for Teflon filters. All reagents were prepared using chemicals supplied by Merck (Analytical grade). Inductively coupled plasma-optical emission spectrometer (ICP-OES, ICP-OES Optima 2100DV, PerkinElmer) was used to analyze the metal concentrations.

Analytical drift was monitored throughout the procedure. Recovery efficiencies were determined and analyzed using a diluted sample spiked with a known quantity of metal. Recovery efficiencies from 93.7 to 100.8% were achieved. The method detection limit (MDL) was estimated by repeatedly analyzing a predefined quality control solution and by replicate analysis in ICP- OES measurements, the MDL of each element was calculated by MDL = 2.681 × S_{pooled}, with $S_A^2/S_B^2 < 3.05$. $S_{pooled} = [(6S_A^2 + 6S_B^2)/12]^{0.5}$, where S_{pooled} is the pooled standard deviation, S_A the standard deviation of the one of two prepared samples with a bigger F-test value, and S_B the standard deviation of the other. The detection limits in ng m^{-3} (calculated from MDL × volume of analyte solution (25 mL)/average sampling volume (40 m^3)) were 16.8, 0.113, 0.719, 0.96, 2.35, 0.264, 0.066, 0.042, 0.154, 0.313, 0.047, 0.033, 0.050, and 0.018 for Na, Mg, Al, K, Ca, Fe, Cr, Ni, Zn, Sr, Ba, Pb, Mn, and Cu, respectively.

RESULTS AND DISCUSSION

Concentrations of Metals in PM with Various Aerodynamic Diameters before and after Beehive Firework Display

Tables 1 and 2 present the concentrations of metals in particles before (background), on the rooftop of the Wumiao Temple (windward) and YanShuei police station (leeward) during and after a beehive firework display as part of the 2013 YanShuei Firework Festival. The mass concentration of $PM_{2.5}$ increased from 28.2 µg m^{-3} (background) to 437 µg m^{-3} during the beehive display on the leeward side, and from 26.1 µg m^{-3} to 165 µg m^{-3} on the windward side. At the windward sampling site, the $PM_{2.5}$ and PM_{10} concentrations during the display reached approximately 4.7 and 4.9 times the background values, respectively, whereas at the leeward site, they were 12.5 and 4.6 times the national standards (35 and 125 µg m^{-3}, respectively). Joly *et al*. (2010) reported that the highest $PM_{2.5}$ levels during the 2007 Montréal International Fireworks Competition were almost 10,000 µg m^{-3}, which is approximately 1,000 times the background level.

Before beehive firework display (background), the total mean concentrations of 14 metals in $PM_{1.0}$, $PM_{2.5}$, and PM_{10} particles were 349, 1016, and 3431 ng m^{-3}, respectively, at the leeward site and 2085, 2971, and 3785 ng m^{-3}, respectively, at the windward site. The major metals (30–100 ng m^{-3}) in $PM_{1.0}$, $PM_{2.5}$, and PM_{10} were Al, Ca, Fe, K, Na, and Zn (crustal metals and the metallic constituents of sea salt). During the displays, the metals that were generated at high concentration (≥ ~10^3 ng m^{-3}) were Al, Ba, Cu, K, Na, and Sr. During the beehive firework display, the concentrations of metals in $PM_{2.5}$ (D) at leeward sampling site rise above the background level (B) to degrees that decrease in the order Ba (1,828 times), K (702 times), Sr (534 times), Cu (473 times), Mg (104 times), Al (121

Table 1. Concentrations of metals in $PM_{1.0}$, $PM_{2.5}$, and PM_{10} at the leeward sampling site.

Particle size	Sampling period	PM ($\mu g\ m^{-3}$)	Metals concentration ($ng\ m^{-3}$)													
			Al	Ba	Ca	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sr	Zn
$PM_{1.0}$	Background	20.9	30.27	0.9719	31.04	2.944	1.615	52.31	87.09	9.822	3.073	65.73	2.873	7.025	2.115	51.89
	Trial	77.9	621.0	48.83	316.9	28.94	21.17	488.3	1637	125.6	20.60	786.6	24.24	74.65	4.094	274.6
	During	321	5761	2951	164.6	44.12	1092	319.4	72977	5830	79.41	1237	4.518	883.1	1163	980.0
	After	43.5	73.79	9.306	2.532	4.726	7.110	102.8	271.3	55.69	5.286	75.91	4.079	22.75	2.990	120.1
$PM_{2.5}$	Background	28.2	62.69	2.082	83.80	4.368	3.006	126.2	126.2	73.36	6.284	400.3	3.241	11.76	2.745	109.5
	Trial	120	1045	58.96	801.9	112.4	23.62	1019	2025	567.3	41.29	1896	29.62	96.11	12.51	517.5
	During	437	7603	3805	462.7	58.31	1421	634.7	88637	7636	110.2	2133	5.559	1150	1466	1249
	After	107	190.8	23.47	108.1	7.028	14.77	286.4	537.7	193.7	16.61	481.8	5.003	52.23	8.391	312.7
PM_{10}	Background	40.6	209.8	4.559	300.7	6.509	16.49	358.1	217.2	313.3	11.92	1716	60.53	40.68	4.601	170.8
	Trial	181	1522	66.67	1550	199.2	24.33	1613	2403	1223	55.46	4962	33.35	111.2	19.10	721.1
	During	572	11001	4601	2134	69.94	1611	1712	98807	9662	150.8	5571	7.984	1198	1596	1405
	After	163	788.6	51.97	889.9	11.21	25.31	983.1	944.6	861.7	35.58	2401	6.364	63.57	20.21	462.8

Table 2. Concentrations of metals in $PM_{1.0}$, $PM_{2.5}$, and PM_{10} at the windward sampling site.

Particle size	Sampling period	PM ($\mu g\ m^{-3}$)	Metals concentration ($ng\ m^{-3}$)													
			Al	Ba	Ca	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sr	Zn
$PM_{1.0}$	Background	17.4	128.6	3.252	393.2	2.159	10.25	161.5	615.6	134.8	43.97	444.0	11.56	20.50	1.893	114.2
	Trial	30.6	54.20	1.803	90.64	2.159	9.840	67.71	154.0	18.63	13.61	225.3	3.808	20.03	1.144	70.03
	During	120	765.3	167.2	698.9	16.32	86.12	211.7	9744	718.1	113.1	408.5	14.37	85.17	115.6	146.3
	After	45.7	81.57	9.026	227.9	4.388	11.70	101.0	977.7	108.9	27.79	87.27	12.47	33.96	3.000	90.58
$PM_{2.5}$	Background	26.1	190.8	4.109	556.3	3.055	12.34	253.9	697.1	212.0	65.27	784.1	14.65	24.93	2.620	149.7
	Trial	46.1	125.5	3.646	194.7	4.737	16.35	181.0	224.1	109.3	19.43	598.0	4.704	27.08	2.609	165.8
	During	165	1011	222.3	1010	19.76	123.9	432.2	8104	1010	142.1	1047	20.15	122.2	140.7	250.2
	After	103	169.4	24.14	398.5	5.713	21.87	259.9	1251	222.5	35.24	570.0	15.61	64.36	7.972	238.3
PM_{10}	Background	45.4	262.3	4.910	731.7	4.237	14.44	374.0	764.2	281.6	84.70	1041	18.98	27.78	3.352	172.1
	Trial	85.4	388.4	7.033	684.9	6.190	22.69	537.0	348.6	461.1	33.22	1864	6.191	30.36	6.013	316.9
	During	222	1746	250.8	1863	26.15	156.0	1064	6775	1801	252.8	4088	32.48	146.8	154.0	334.4
	After	148	475.6	33.82	1063	7.829	28.12	706.4	1463	613.9	53.94	2352	20.15	77.39	13.68	316.3

times), and Pb (98 times). The corresponding data (D/B) for $PM_{1.0}$ were Ba (3,036 times), K (838 times), Cu (676 times), Mg (594 times), Sr (550 times), Al (190 times), and Pb (126 times). In recent years, demand for fireworks of multiple colors has considerably increased the use of metals as color developers. Accordingly, during the beehive firework display, the mass concentrations of Ba (green coloring agent), Sr (red), Cu (blue) (Kulshrestha *et al*., 2004; Wang *et al*., 2007; Moreno *et al*., 2007; Perrino *et al*., 2011) in $PM_{2.5}$ at the leeward sampling site increased above their background values (Ba: 2.082; Sr: 2.745; Cu: 3.006 ng m^{-3}) by 1828, 534 and 473 times, respectively (to Ba: 3,805; Sr: 1,466; Cu: 1,421 ng m^{-3}) .

At the windward sampling site, the D/B values in $PM_{2.5}$ were, in decreasing order, Sr (×74), Ba (×68), K (×13), and Cu (×12). The concentrations of Ba, K, Cu, Mg, and Sr in the particulates were significantly higher at the leeward site than at the windward site. Most investigations of atmospheric PM emissions from fireworks have focused on Sr, Ba, and K as tracers of firework emissions (Liu *et al*., 1997; Kulshrestha *et al*., 2004; Drewnick *et al*., 2006; Moreno *et al*., 2007; Wang *et al*., 2007; Barman *et al*., 2008; Vecchi *et al*., 2008; Galindo *et al*., 2009; Joly *et al*., 2010). Barium compounds ($BaClO_3$ and $Ba(NO_3)_2$) are used as oxidizers. Both $BaCO_3$ and $Ba(NO_3)_2$ are used to create white effects or, in the presence of chlorine, bright green a firework color that is mostly associated with Ba (Lancaster *et al*., 1998; Perry, 1999). Potassium compounds (KNO_3, $KClO_4$, and $KClO_3$), which are used as propellants in fireworks, are the main oxidizers: in a firework display. Lin *et al*. (2014) found that the concentrations of Cl^- in $PM_{1.0}$, $PM_{2.5}$, and $PM_{2.5-10}$ during a display were 91, 64, and 6.9 times higher than their background values. Furthermore, increases in measured K concentrations suggest that $KClO_3$ and $KClO_4$ are the major sources of oxygen in firecrackers. Both $SrSO_4$ and $Sr(NO_3)_2$ can be used as oxidizers, and, along with carbonate, impart a red color to fireworks when combined with chlorine. Finally, copper compounds such as the copper chloride and copper oxide produce a blue color, and can be mixed with strontium compounds to produce purple effects. $CuCr_2O_4$ is used as a catalyst in rocket propellants (Lancaster *et al*., 1998). Although K, as a black powder fuel that is combined with S, dominates "special effect"trace additives can include various other metals such as Al, Cu, Ti, or even Pb (Hickey *et al*., 2010). Pb is of particular interest, given its high toxicity, as it is one of the few metals/metalloids for which legal atmospheric concentration limits exist (along with As, Hg, Ni, and Cd). Nevertheless, in many countries, laws against the use of Pb in the manufacture and combustion of fireworks are being thwarted by imports from countries that are less concerned with the potential health implications of their products. Some fireworks continue to contain Pb levels that are measurable in decigrams (Hickey *et al*., 2010). During a beehive firework display, the total Pb in PM_{10} was 1,198 ng m^{-3} — higher than the air quality limit for Pb (500 ng m^{-3}) that has been set by the World Health Organization (WHO, 2000). In the present investigation, the Pb concentrations in fine particles (1,150 ng m^{-3}) and $PM_{1.0}$ (883.1 ng m^{-3}) were found to be 24 and 18 times those in coarse particles. Therefore, the short-term exposure to firework sources should be concerned for adverse health effects because it is easier for fine-particulate Pb to enter and accumulate in the human respiratory system than for coarse-particulate Pb.

Ratios (T/B, D/B, and A/B) (Ratios of Values of Particle-Bound Metal Concentrations for Various PMs Before (Background (B)), Trail (T), During (D), and After (A) Beehive Firework Display of Particle-Bound metal Compositions for Different PMs

According to Fig. 1, the D/B ratios of particle-bound Ba, Cu, K, Mg, and Sr in $PM_{1.0}$ and $PM_{2.5}$ at the leeward sampling site were significantly higher than T/B and A/B during the beehive firework display. The maximal D/B values of Ba were 198 and 118 in $PM_{1.0}$ and $PM_{2.5}$ during the beehive fireworks display at that site. The D/B values for $PM_{1.0}$ and $PM_{2.5}$ were lower at the windward site. The maximal D/B value of Ba was 8.56 for $PM_{2.5}$ and the maximal D/B value of Sr was 8.85 for $PM_{1.0}$. The D/B values of particle-bound Ca, Cr, Fe, Mn, Na, Ni, and Zn in $PM_{1.0}$ and $PM_{2.5}$ were less than corresponding T/B and A/B values at both leeward and windward sites. (Notably, the ratios for the metals compositions were all between zero and two.) This finding suggests that concentrations of particle-bound Al, Ba, Cu, K, Mg, Pb, and Sr increased during the beehive firework display, suggesting that firework aerosols are rich in Al, Ba, Cu, K, Mg, Pb, and Sr.

The above inference was examined by comparing the metal concentration distributions normalized to the concentrations of the upper continental crust (UCC) (Weckwerth, 2001) for the differently sized particles at the leeward and windward sampling sites during the sampling period (Fig. 2). The sampled $PM_{1.0}$, $PM_{2.5}$, and PM_{10} particles all exhibited similar distributions of concentrations of the crustal metals (Al, Ca, Fe, Mg, and Na), normalized to UCC, for the trial, during, and after a beehive fireworks display at the leeward and windward sites; furthermore, these patterns were also similar to the background. Interestingly, the relative abundances of Ba, Cu, K, Pb, and Sr in $PM_{1.0}$ and $PM_{2.5}$ during the beehive firework display at the leeward site were very different to the background abundances, indicating that the beehive firework display emited more of these five metals in $PM_{1.0}$ and $PM_{2.5}$. The concentrations of these five metals in $PM_{1.0}$ and $PM_{2.5}$, relative to those in the UCC, in the trial, during, and after the beehive firework display at the windward site were close to the background values.

Size Distributions of Particulate Metals before and after the Beehive Firework Display

Fig. 3 presents the size distribution of particles of the major metals (Ba, K, Cu, Mg, Pb, and Sr) during four periods during the sampling period (February 21^{st}–22^{nd}, 23^{th}–24^{th}, 24^{th} (18:00–24:00), and 25^{th}, 2013 , which were before (background), trial, during, and after the beehive firework display, respectively). The results in the figure demonstrate that, at the leeward sampling site, the major metals exhibited approximately bi-modal size distributions with primary peaks in the 0.56–1.0 μm range and secondary peaks in the

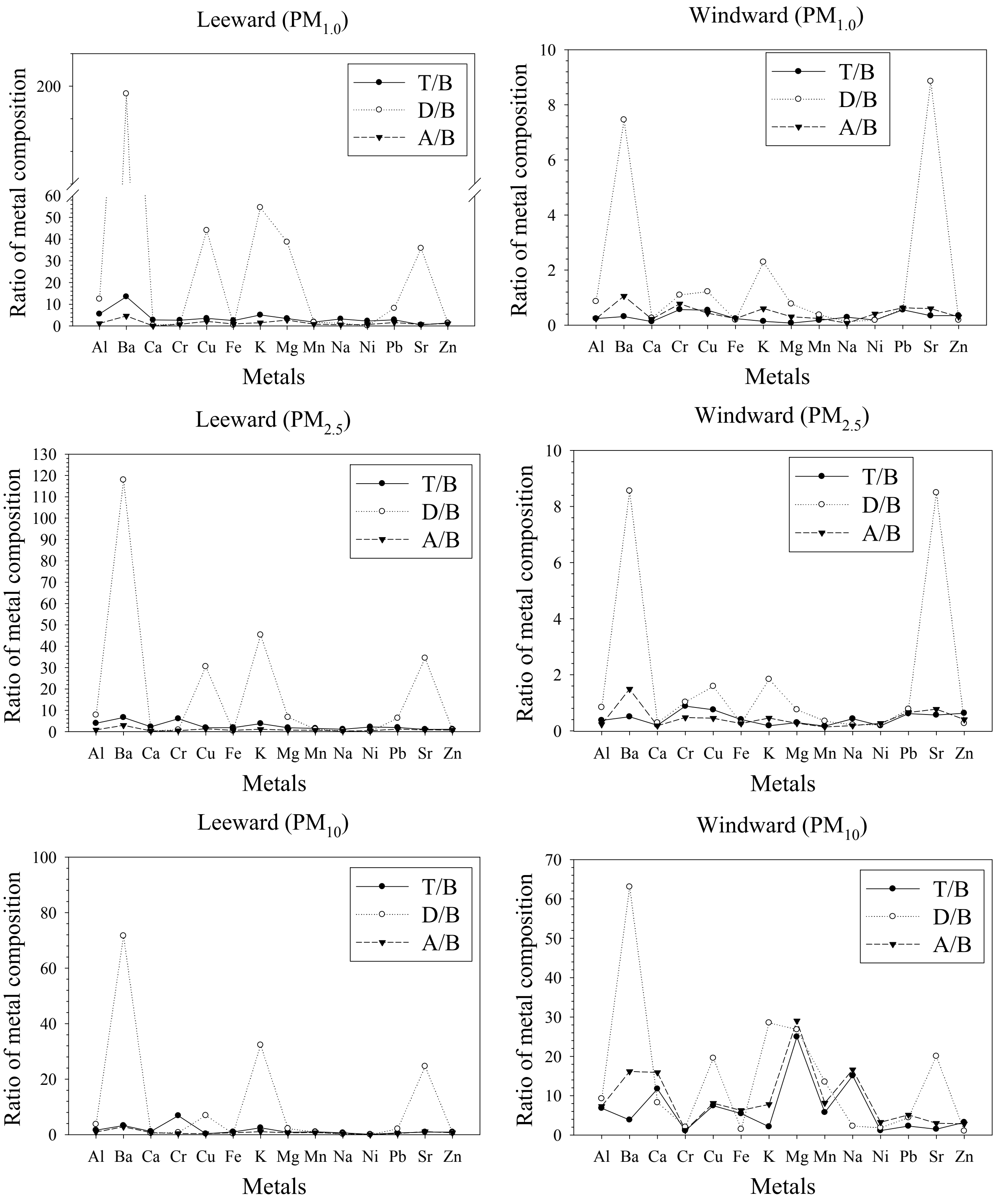

Fig. 1. T/B, D/B, and A/B values and ratios of compositions of particle-bound metals in different particle size ranges at the leeward and windward sampling sites (B: background; D: during beehive fireworks display; A: after the display).

coarse size range (3.2–5.6 µm) during the beehive firework display. At the leeward sampling site, before the beehive firework display (Background), the concentration of K exhibited a bi-modal size distribution, but those of the other major metals exhibited approximately single-modal size distributions with major peaks in the coarse particle range (3.2–5.6 µm). These distributions differed greatly from those during the beehive firework display. The major peaks in the size distribution of the metal particles clearly shifted from coarse particles to fine particles during the beehive firework display at the leeward site.

Fig. 4 displays cumulative mass fractions of the particle-bound metals at the leeward and windward sampling sites. At the leeward sampling site, the ranges of mass ratios of the major metals to $PM_{2.5}$ and PM_1 were 19.0–55.3% and 2.8–43%, respectively, for the background and 74.5–95.3% and 56.5–72.9%, respectively, during the beehive firework display. At the leeward sampling site, the percentages of major metals (Ba, K, Cu, Mg, Pb, and Sr) in PM_1 and $PM_{2.5}$ during the beehive firework display exceeded the background value. Ba, K, Cu, Mg, Pb, and Sr, commonly regarded as firework display indicator elements (Vecchi *et al.*, 2008; Joly *et al.*, 2010; Crespo *et al.*, 2012; Tsai *et al.*, 2012; Kumar, *et al.*, 2016), accounted for 0.520, 2.45, 26.4 and 0.849%, respectively, of the mass of PM_1, and 0.777, 2.32, 23.8, and 0.776%, respectively, of the mass of $PM_{2.5}$ before (background), trial, during, and after the beehive firework display. Therefore, the mass ratios of these metals during the display were higher than the background values, trial, and the values of after the display.

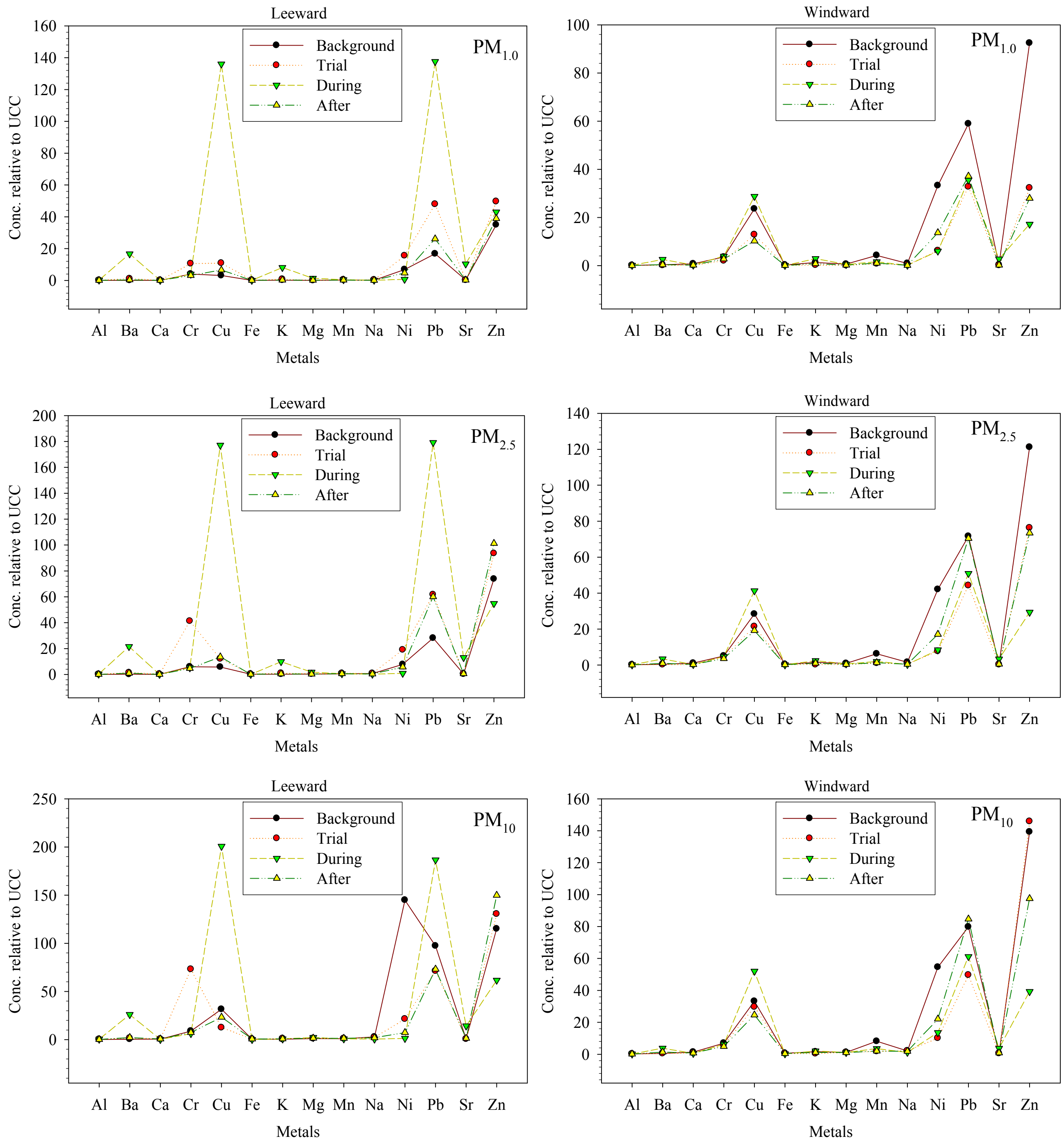

Fig. 2. Metals distribution patterns (normalized to upper continental crust (UCC)) for the sized particles at the leeward and windward sampling sites.

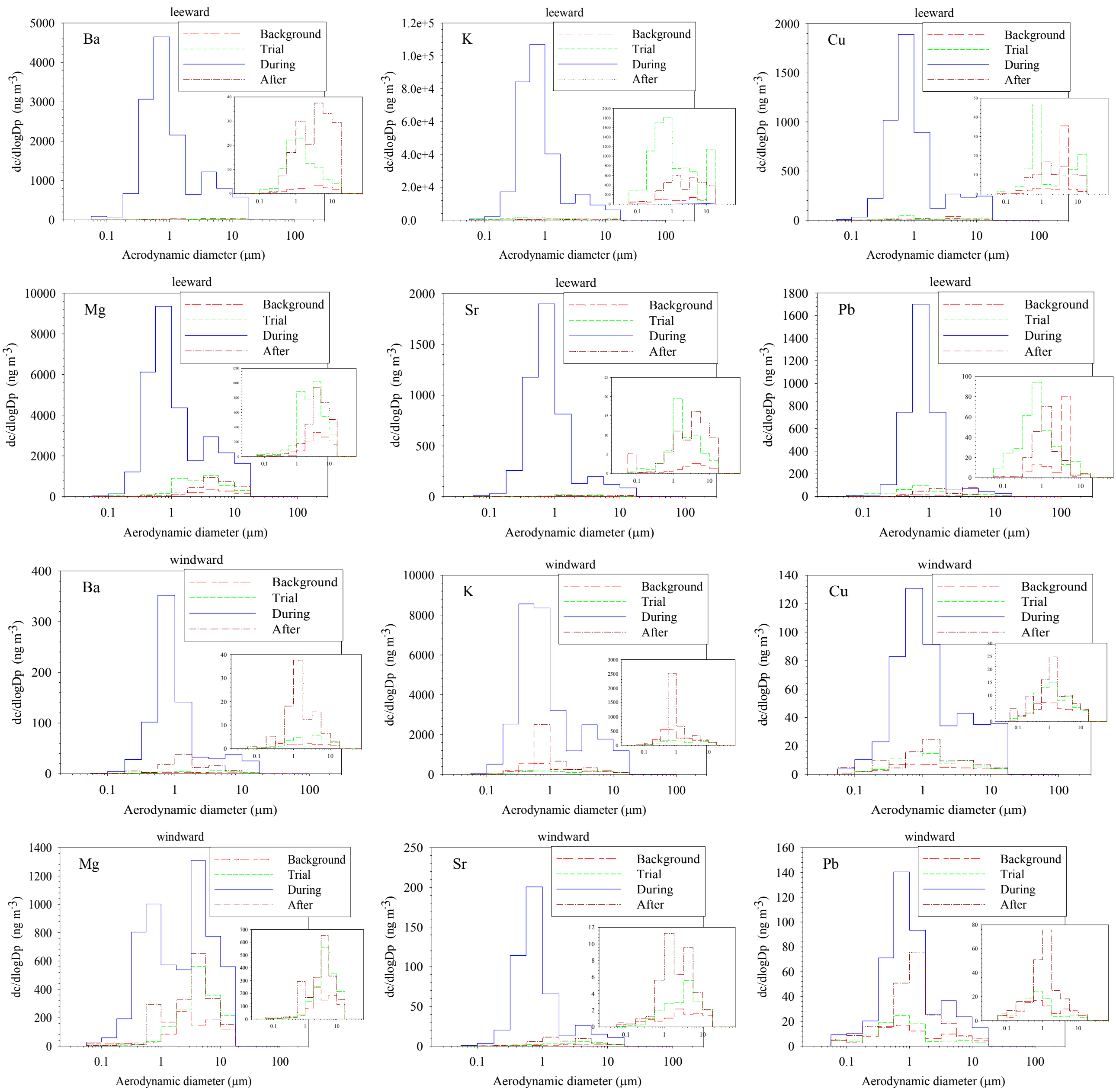

Fig. 3. Size distributions of the particle-bound major metals at the leeward and windward sampling sites.

Analysis of Particulate Metal Sources

Metal species with characteristic values of over 1 in Principal Component Analysis (PCA) (SPSS v.12.0) can be classified into several groups by their sources (Allen *et al.*, 2001; Marcazzan *et al.*, 2001; Manoli *et al.*, 2002; Al-Momani, 2003; Azid *et al.*, 2015; Chen *et al.*, 2015; Fang *et al.*, 2015; Liang *et al.*, 2015). In each sampling period, the metal species, based on their characteristic values, exhibited three groups in PCA (Table 3). For the metal species in $PM_{2.5}$ before the beehive firework display, these three groups had characteristic values of 6.06, 5.68, and 1.25, corresponding to the potential sources of vehicles, resuspended dust, and wear of brake-linings/tires, respectively. The characteristic values in the particles during the trial beehive firework displays were 6.72, 3.85, and 1.44, revealing that their sources were fireworks, vehicles, and fuel oil, respectively. The fine particles also had three groups with characteristic values of 8.30, 4.06, and 1.22, referring to the sources of fireworks, resuspended dust, and fuel oil, respectively. The assessments of the firework source chemical profile and the effect of the fireworks display on the local environment represent an original contribution toward a better understanding of the aerosol characteristics and burdens during firework displays. After the beehive fireworks display, the fine particles had group characteristic values of 5.70, 5.40, and 1.17, corresponding to likely sources of resuspended dust, fireworks, and industry, respectively. The PCA analysis also revealed that the dominant metal elements of fine particles during fireworks were Al, Ba, Cr, Cu, K, Mg, Mn, Pb, Sr, and Zn.

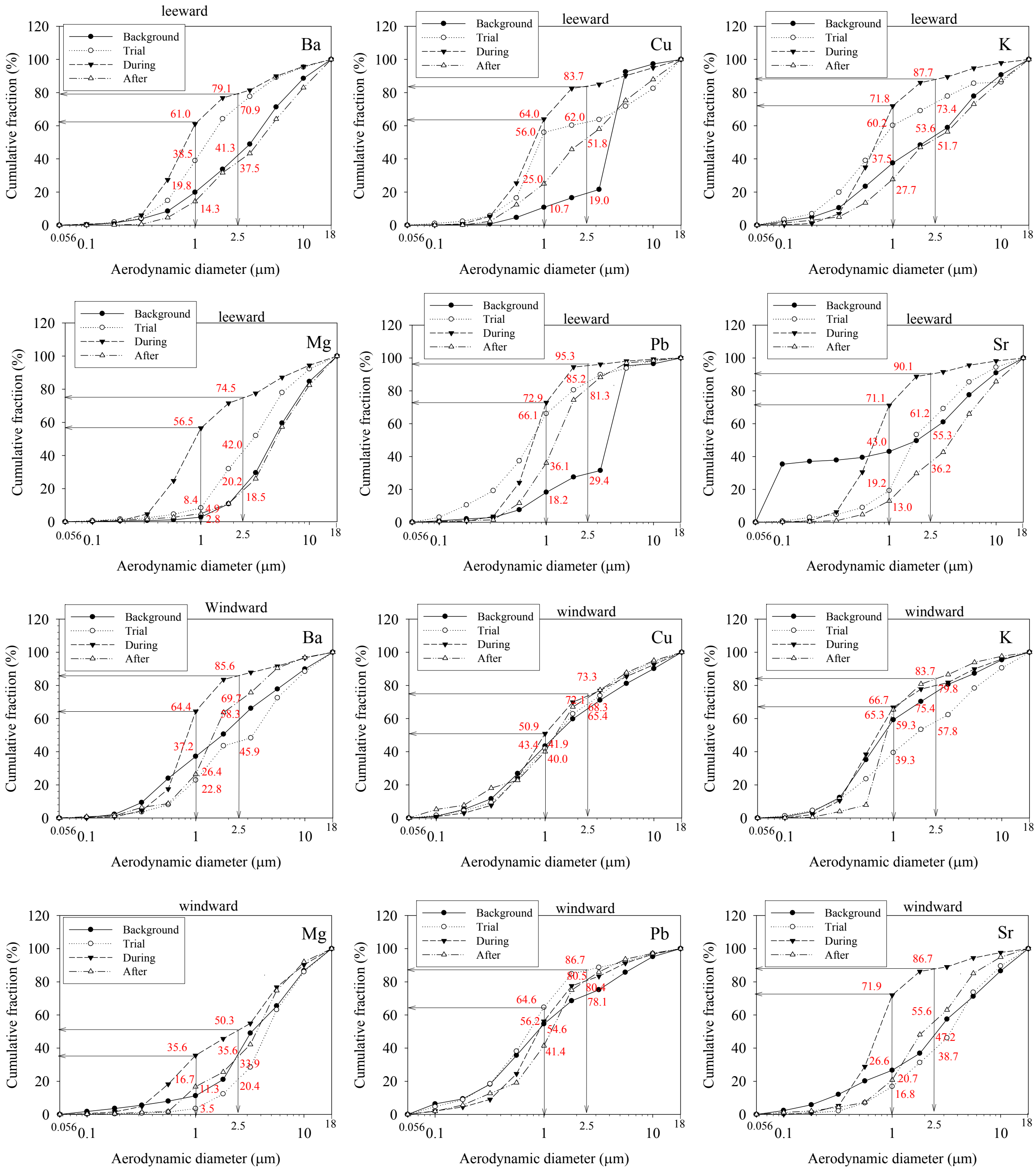

Fig. 4. Cumulative mass fractions of the particle-bound major metals at the leeward and windward sampling sites.

CONCLUSIONS

In this study, the samples of metals in sized particles were collected using a MOUDI sampler in the YanShuei area of southern Taiwan during a beehive firework display. The results indicate that during the beehive firework display, the ratios of metal concentrations in $PM_{2.5}$ (D) to the background level (B) at leeward sampling site were 1,828 for Ba, 702 for K, 534 for Sr, 473 for Cu, 104 for Mg, 121 for Al, and 98 for Pb. The corresponding data for $PM_{1.0}$ were 3036, 838, 550, 676, 594, 190, and 126, respectively. According to the analyses of metal composition ratio, PCA, and UCC, the concentrations of particle-bound Al, Ba, Cu, K, Mg, Pb, and Sr increased during the beehive firework displays, suggesting that firework-display aerosols contained abundant metal elements of Al, Ba, Cu, K, Mg,

Table 3. Principal component analysis for the metals in $PM_{2.5}$ during the four sampling periods.

Element	Background			Trail			During			After		
	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3
Al		0.89		0.69	0.52		0.69	0.65		0.91		
Ba		0.88		0.53	0.60		0.95			0.46	0.85	
Ca	0.90			0.98				1.00		0.96		
Cr	0.78	0.54					0.92					0.64
Cu	0.98					0.69	0.99				0.97	
Fe		0.98		0.97				0.99		0.90		
K	0.80				0.96		0.96				0.54	
Mg		1.00		0.99			0.91			0.88		
Mn	0.93			0.87			0.73	0.63		0.71		
Na		0.97		0.86				0.99		0.92		
Ni	0.97				0.97				0.98		0.91	
Pb	0.87				0.89		0.95				0.98	
Sr		0.87		0.91			0.99			0.63	0.75	
Zn			0.94	0.82			0.95				0.85	
Eigenvalues	6.06	5.68	1.25	6.72	3.85	1.44	8.30	4.06	1.22	5.70	5.40	1.17
Variance %	43.3	40.6	8.92	48.0	27.5	10.3	59.3	29.0	8.73	40.7	38.6	8.37
Cumulative %	43.3	83.8	92.8	48.0	75.5	85.8	59.3	88.3	97.0	40.7	79.3	87.7
Source types	V	R	B/T	F	V	O	F	R	O	R	F	I

B/T: Brake-lining/Tire rubbed off, V: Vehicles, F: Fireworks, I: Industry, O: Oil, and R: Resuspended dust.

Pb, and Sr. The Pb concentrations in fine particles (1,150 ng m^{-3}) and $PM_{1.0}$ (883.1 ng m^{-3}) were 24 and 18 times those in coarse particles, respectively. Therefore, the short-term exposure to firework sources should be concerned for adverse health effects because it is easier for fine-particulate Pb to enter and accumulate in the human respiratory system than for coarse-particulate Pb.

The major peaks in the size distribution of the metal particles clearly shifted from coarse particles to fine particles during the beehive firework display at the leeward site. Before (background), trial, during, and after the beehive firework display, the Ba, K, Cu, Mg, Pb, and Sr (commonly regarded as firework display indicator elements) in PM_1 accounted for 0.520, 2.45, 26.4 and 0.849% of PM mass respectively, while in $PM_{2.5}$ the corresponding values were 0.777, 2.32, 23.8, and 0.776%, respectively.

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