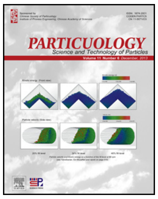
Particuology xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Particuology

journal homepage: www.elsevier.com/locate/partic



$PM_{2.5}$ and $PM_{10\text{-}2.5}$ chemical composition and source apportionment near a Hong Kong roadway☆

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ARTICLE INFO

Article history:
Received 7 August 2013
Received in revised form 24 October 2013
Accepted 29 October 2013

Keywords:
$PM_{2.5}$
$PM_{10\text{-}2.5}$
Roadside
Chemical composition
Source apportionment
Hong Kong

ABSTRACT

Twenty-four-hour $PM_{2.5}$ and PM_{10} samples were collected simultaneously at a highly trafficked roadside site in Hong Kong every sixth day from October 2004 to September 2005. The mass concentrations of $PM_{2.5}$, $PM_{10\text{-}2.5}$ (defined as $PM_{10} - PM_{2.5}$), organic carbon (OC), elemental carbon (EC), water-soluble ions, and up to 25 elements were determined. Investigation of the chemical compositions and potential sources revealed distinct differences between $PM_{2.5}$ and $PM_{10\text{-}2.5}$. The annual average mass concentrations were 55.5 ± 25.5 and 25.9 ± 15.7 μg/m^3 for $PM_{2.5}$ and $PM_{10\text{-}2.5}$, respectively. EC, OM ($OM = OC \times 1.4$), and ammonium sulfate comprised over ~82% of $PM_{2.5}$, accounting for ~29%, ~27%, and ~25%, respectively, of the $PM_{2.5}$ mass. Low OC/EC ratios (less than 1) for $PM_{2.5}$ suggested that fresh diesel-engine exhaust was a major contributor. Seven sources were resolved for $PM_{2.5}$ by positive matrix factorization (PMF) model, including vehicle emissions (~29%), secondary inorganic aerosols (~27%), waste incinerator/biomass burning (~23%), residual oil combustion (~10%), marine aerosols (~6%), industrial exhaust (~4%), and resuspended road dust (~1%). EC and OM comprised only ~19% of $PM_{10\text{-}2.5}$. The average OC/EC ratio of $PM_{10\text{-}2.5}$ was 7.8 ± 14.2, suggesting that sources other than vehicular exhaust were important contributors. The sources for $PM_{10\text{-}2.5}$ determined by the PMF model included ~20% traffic-generated resuspension (e.g., tire dust/brake linear/petrol evaporation), ~17% locally resuspended road dust, ~17% marine aerosols, ~12% secondary aerosols/field burning, and ~11% vehicle emissions.

1. Introduction

On-road vehicles are a major source of air pollution in the megacities of the world (Mage et al., 1996). As environmental and health awareness increases in Hong Kong, concerns about particulate matter (PM) pollution are also increasing due to the continuously growing vehicle numbers and kilometers driven. Emission inventories in Hong Kong show that vehicle emissions account for ~30% of PM_{10} (http://www.epd.gov.hk). Approximately ~30% of all vehicles in Hong Kong utilize diesel fuel; thus, street-level human exposure to vehicle emissions is of concern.

Emissions from on-road vehicles have a wide range of particle sizes (Thorpe & Harrison, 2008) from ultrafine and fine to coarse (particles with aerodynamic diameters of <100 nm, <2.5 μm, and between 10 and 2.5 μm, respectively). $PM_{2.5}$ and $PM_{10\text{-}2.5}$ particles have different sources, chemical compositions, and removal processes, even in the same urban traffic environment (Li, Wiedinmyer, & Hannigan, 2013). $PM_{2.5}$ mainly originates from combustion sources, such as engine exhausts (Cheng, Lee, et al., 2010; Ho, Cao, Lee, & Chan, 2006; Li, Wiedinmyer, et al., 2013; Toner, Sodeman, & Prather, 2006) and biomass burning (Cheng, Liu, & Chen, 2010; Fraser, Cass, & Simoneit, 1998). $PM_{10\text{-}2.5}$ is predominantly due to resuspended road dust (Abu-Allaban,

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1674-2001/$ – see front matter © 2013 Published by Elsevier B.V. on behalf of Chinese Society of Particuology and Institute of Process Engineering, Chinese Academy of Sciences.
http://dx.doi.org/10.1016/j.partic.2013.10.003

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Gillies, Gertler, Clayton, & Proffitt, 2003; Kupiainen et al., 2005; Li, Wiedinmyer, et al., 2013) and mechanical processes, such as tire and brake emissions (Councell, Duckenfield, Landa, & Callender, 2004). Harrison et al. (2000) considered the coarse particle fraction to consist of two components, a nonwind-suspended component from anthropogenic sources, such as industrial processes, construction activities, traffic-generated resuspension, and biological particles, and a wind-suspended component mainly from natural sources, such as sea spray and surface soils or dusts on paved areas. Differences in the chemical characteristics and evidence from epidemiological studies of adverse health effects indicate that separate standards should be set for coarse particles. A recent study in Hong Kong revealed that $PM_{10\text{-}2.5}$ was associated with emergency hospital admissions for respiratory diseases independent of $PM_{2.5}$ and gaseous pollutants (Qiu et al., 2012).

Roadside air quality studies of real-world emissions are limited in Hong Kong, although elevated PM mass concentrations have been attributed to vehicle engine emissions in many urban atmosphere, such as Hong Kong (Ho et al., 2006), Beijing (Ma et al., 2011), Xi'an (Cao et al., 2005), Shanghai (Waheed et al., 2010), Guangzhou (Wang, Bi, Sheng, & Fu, 2006), and cities in the United States (Li, Wiedinmyer, et al., 2013). Roadside $PM_{2.5}$ studies in Hong Kong have mainly focused on the spatial variations instead of the source characteristics (e.g., Ho et al., 2006; Louie, Chow, et al., 2005; So, Guo, & Li, 2007). In this study, $PM_{2.5}$ and $PM_{10\text{-}2.5}$ samples collected over 24 h at a roadside sampling site are investigated to (1) determine the chemical characteristics of $PM_{2.5}$ and $PM_{10\text{-}2.5}$ and (2) evaluate the source attributes in a vehicle-dominated urban environment.

2. Methods

2.1. Sampling location

The climate in Hong Kong is subtropical and influenced by the Asian monsoons. The cooling and heating of the Asian land mass during winter and summer, respectively, give rise to large-scale monsoonal winds, resulting in prevailing winds from the north/northeast/east during cold seasons (September to April) and southwest during the warm seasons (May to August).

The Hong Kong Polytechnic University (PU) roadside sampling site (22.30° N, 114.17° E) is located at ground level within ~1 m of Hong Chong Road, ~10 m from an eight-floor university building, and ~400 m from a tunnel entrance. The Hong Chong Road, which is a ~30 m wide, four-lane, two-direction road, is the main access to the highly trafficked Cross Harbor Tunnel (CHT). Vehicle classes and numbers are recorded at the toll gates. Near the sampling site, the high traffic flow, which results in frequent braking and congestion, is due to its close proximity to public bus stations (100 m to the southwest), toll gates (200 m to the southeast), and an intersection (200 m to the southwest).

Data from the toll gates provided vehicle counts for nine vehicle classes, including private car, motorcycle, taxi, private/public light bus, private/public single-deck bus, private/public double-deck bus, and light (≤5.5 tons), medium (5.5–24 tons), and heavy-duty (≥24 tons) cargo trucks. The private cars and motorcycles were fueled with gasoline, >99% of the taxis were fueled with liquid petroleum gasoline (LPG), and the buses and cargo trucks were mostly fueled with diesel. During the sampling period, Hong Chong Road had an average traffic flow of 5125 vehicles per hour (ranging from 1030 to 6876 vehicles per hour) with an average of 37.2% gasoline-fueled vehicles (ranging from 10.1% to 68.1%), 31.8% diesel-fueled vehicles (ranging from 12.7% to 54.9%), and 30.7% LPG-fueled taxis (ranging from 7.5% to 69.7%).

2.2. Sample collection

$PM_{2.5}$ and PM_{10} were sampled over a 24-h period every sixth day for one year from 8 October 2004 through 23 September 2005, and 40 valid sample sets were obtained. An integrated filter sampler (Model URG3000 ABC, URG Corporation, Chapel Hill, United States) equipped with three inlets ($PM_{1.0}$, $PM_{2.5}$, and PM_{10} Teflon®-coated aluminum cyclones, URG Corporation, Chapel Hill, United States) was used. The sampling inlets were located approximately 1.5 m above street level. Two parallel channels, each operated at 8.3 L/min with 47 mm Teflon-membrane and quartz-fiber filters, were used for $PM_{2.5}$ and PM_{10}. Both the Teflon-membrane and quartz-fiber filters were weighed twice before and after sampling using a Sartorius model MC5 microbalance (Göttingen, Germany) with a sensitivity of ±1 μg in the 0–250 mg range. Before weighing, the filters were equilibrated for 24 h in a desiccator at a temperature of 20–30 °C and relative humidity of 30–40%. Prior to sampling, the quartz-fiber filters were preheated in an electric furnace at 900 °C for 3 h to remove carbonaceous contaminants. After sampling, the collected quartz-fiber samples were stored in air-tight containers in a refrigerator at approximately 4 °C to prevent the evaporation of volatile components prior to the chemical analyses.

2.3. Chemical analysis

After the gravimetric analysis, the Teflon-membrane filters were analyzed at the Environmental Analysis Facility of the Desert Research Institute (DRI, Reno, NV, United States) for 51 elements (from Na to U) by X-ray fluorescence (PAN Analytical, Netherlands) following the methodology described in Watson, Chow, and Frazier (1999). Half of the quartz-fiber filters were extracted with distilled deionized water and analyzed for chloride (Cl^-), nitrate (NO_3^-), sulfate (SO_4^{2-}), water-soluble sodium (Na^+), potassium (K^+), calcium (Ca^{2+}), and ammonium (NH_4^+) ions by ion chromatography (DIONEX 600, United States) following the standard methodology (Chow & Watson, 1999) in the Air Laboratory of the Hong Kong Polytechnic University. OC and EC were measured on 0.5 cm^2 quartz-fiber filter punches from the remaining half of the quartz-fiber filters using a DRI Model 2001 carbon analyzer (Atmoslytic Inc., Calabasas, CA) and thermal/optical reflectance (TOR) method following the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol (Chow et al., 1993, 2005, 2007, 2011; Chow, Watson, Chen, Arnott, & Moosmuller, 2004; Chow, Watson, Crow, Lowenthal, & Merrifield, 2001). Replicate analyses were performed for ~10% of all samples. Approximately 5% of the field blanks were collected to subtract passive adsorption/deposition and error propagation.

Major mass constituents, including OC, EC, SO_4^{2-}, NO_3^-, NH_4^+, and some elements (e.g., Al, Si, Ca, Fe, and Zn), were detected in almost every sample. For the XRF analysis, the concentrations of only 25 elements were reported because elements such as Sc, Co, Ga, Se, Y, Nb, Mo, Pd, Ag, Cd, In, Sb, Cs, La, Ce, Sm, Eu, Tb, Hf, Ta, Wo, Ir, Au, Hg, Tl, and U were seldom detected in concentrations higher than three times their respective minimum detectable limits.

3. Results and discussion

3.1. $PM_{2.5}$ and $PM_{10\text{-}2.5}$ mass concentrations and mass closures

Gravimetric mass concentrations acquired from the Teflon-membrane and quartz-fiber samples were compared for $PM_{2.5}$ and PM_{10}. As shown in Fig. 1, good agreement (slopes close to 1) and high correlations (correlation coefficients $R^2 > 0.95$; $n = 40$)

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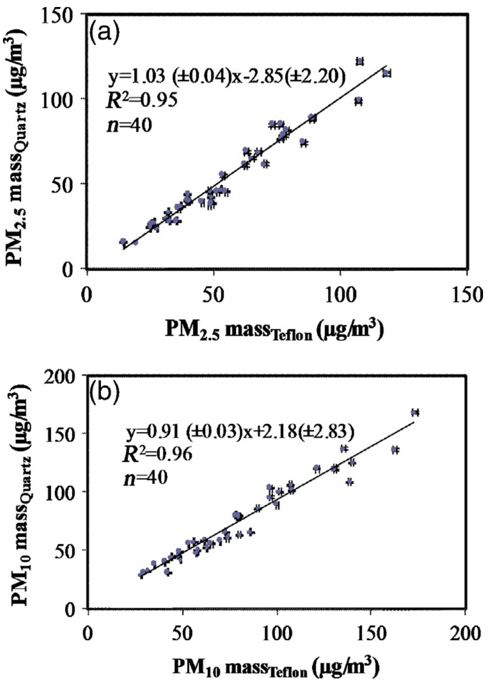

Fig. 1. Comparison of gravimetric mass concentrations from Teflon-membrane and quartz-fiber filters.

between the Teflon-membrane and quartz-filter results indicated consistent sampling and gravimetrical analysis. A similar phenomenon was observed by Engelbrecht, Swanepoel, Chow, Watson, and Egami (2001) in the United States and Louie, Chow, et al. (2005) in Hong Kong. Because quartz-fiber filters are hygroscopic, exhibit positive sampling artifacts due to the absorption of gaseous organic compounds, and tend to shred and suffer material loss after heating, the following discussion refers to the Teflon-membrane mass unless otherwise specified.

The annual average mass concentrations were 55.5 ± 25.5 μg/m^3 for $PM_{2.5}$ and 25.9 ± 15.7 μg/m^3 for $PM_{10\text{-}2.5}$ (Table 1). $PM_{1.0}$ was investigated during the same sampling period, and the results were reported elsewhere (Cheng et al., 2011). On average, $PM_{1.0}$ accounted for ~80% of $PM_{2.5}$, $PM_{2.5}$ comprised ~70% of PM_{10}, and $PM_{10\text{-}2.5}$ accounted for nearly 30% of PM_{10}. These levels were similar to the concentrations of $PM_{2.5}$ and $PM_{10\text{-}2.5}$ (56.8 ± 16.0 and 19.3 ± 26.8 μg/m^3, respectively) measured by Chan and Kwok (2001) in a nearby roadside study during 1998–1999. The smaller $PM_{10\text{-}2.5}$ mass concentration in Chan and Kwok's study might be due to the further distance of the sampling point (8 m vs. 1 m in this study) from the traffic road. In this study, the $PM_{2.5}$ annual average was similar to that measured during 2000–2001 at a Mong Kok roadside site (Louie, Chow, et al., 2005) but 2.3 times higher than that at the Hok Tsui background site in Hong Kong (Louie, Chow, et al., 2005), and it exceeded the US EPA's annual $PM_{2.5}$ NAAQS of 15 μg/m^3 by a factor of 3.7. The $PM_{2.5}$ and PM_{10} levels were higher than those at most traffic sites in Mediterranean urban areas (Grivas, Chaloulakou, & Kassomenos, 2008) and the United States (Li, Wiedinmyer, et al., 2013), which might be attributed to a higher percentage of diesel-fueled vehicles (~31.8%) on the road in this study. More than 70% of the 24-h $PM_{2.5}$ concentrations were higher than the short-term 24-h NAAQS of 35 μg/m^3 in the United States. Less than 5% of PM_{10} exceeded 180 μg/m^3, the short-term 24-h air quality standard in Hong Kong. All evidence showed that the PU roadside site represents pedestrian exposure in a source-dominated environment (i.e., on-road vehicle exhausts).

The chemical characterization of and seasonal variation in $PM_{2.5}$ were similar to those determined in previous roadside studies (e.g., Louie, Chow, et al., 2005; So et al., 2007). Overall, EC, OM (OM = OC × 1.4), and $(NH_4)_2SO_4$ comprised the majority of $PM_{2.5}$, accounting for ~29%, ~27%, and ~25%, respectively, of the $PM_{2.5}$ mass (Fig. 2). NH_4NO_3, sea salts, geological material, trace elements, and unidentified material only contributed approximately 19% to $PM_{2.5}$. The $PM_{10\text{-}2.5}$ chemical composition, however, was found to be different (Fig. 2), with geological material and trace elements being the most abundant components (~28%), followed by unidentified material (~26%) and OM (~12%). The percentages of sea salt (~10%) and NH_4NO_3 (~10%) were much higher in $PM_{10\text{-}2.5}$ than in $PM_{2.5}$. EC and $(NH_4)_2SO_4$, the major components in $PM_{2.5}$, accounted for only ~7% each of $PM_{10\text{-}2.5}$. The annual average concentrations for the major chemical species in $PM_{10\text{-}2.5}$ followed the decreasing sequence of OC (2.1 ± 1.8 μg/m^3), NO_3^- (1.9 ± 1.6 μg/m^3), EC (1.7 ± 2.7 μg/m^3), SO_4^{2-} (1.4 ± 1.5 μg/m^3), Si (1.3 ± 1.0 μg/m^3), Fe (1.3 ± 0.53 μg/m^3), Cl^- (0.96 ± 0.64 μg/m^3), Na^+ (1.0 ± 0.75 μg/m^3), Al (0.69 ± 0.55 μg/m^3), and Ca (0.77 ± 0.46 μg/m^3) as shown in Table 1.

Several studies also observed a large (20–50%) unidentified mass in $PM_{10\text{-}2.5}$ (Cheung et al., 2011; Hueglin et al., 2005; Yin et al., 2005). This mass might be attributed to the unmeasured water, metal, oxide, hydrogen, and oxygen contents related to geological materials and organics. The unidentified mass can be regressed upon each individual chemical component to identify the related sources (Harrison, Jones, & Lawrence, 2003). The relationships between the unidentified mass and SO_4^{2-} and NO_3^- can be used as an indicator of particle bond water because approximately 80% of particle bond water has been found to associate with SO_4^{2-} and NO_3^-, while 20% associates with organic compounds (Speer, Edney, & Kleindienst, 2003). However, poor correlations ($0.06 < R^2 < 0.09$) between the unidentified mass and ionic species (i.e., $SO_4^{2-}$and NO_3^-) were found in this study.

A poor relationship between OM and the unidentified material with $R^2 = 0.48$ was observed in this study. WSOC (>2.1 μm) largely consisted of the low molecular weight polar compounds, which likely associate with additional water and sea salts. Additional uncertainties in the $PM_{10\text{-}2.5}$ mass closure might be attributed to the factor of 1.4 used to convert OC to OM. A conversion factor of 2.1 was suggested by Chen and Yu (2007) for a suburban site in Hong Kong. In the southeastern United States, Edgerton et al. (2009) reported upper limits of the $PM_{10\text{-}2.5}$ OM/OC conversion factor of 2 for urban sites and 2.6–2.7 for rural sites.

3.2. *Characteristics of the chemical species in $PM_{2.5}$ and $PM_{10\text{-}2.5}$*

The sources and formation of the major chemical constituents were evaluated using the OC/EC ratio and linear regression. OC, EC, SO_4^{2-}, NH_4^+, and K^+ were more abundant in the $PM_{2.5}$ fraction, whereas Cl^-, Al, Si, Ca, Ti, and Fe were enriched in the $PM_{10\text{-}2.5}$ fraction. The ratio of OC to EC has been used to study the emissions and transformation characteristics of carbonaceous aerosols. The underlying hypothesis is that EC originates from primary anthropogenic sources and is not formed by reactions involving gaseous hydrocarbon precursors in the atmosphere. In contrast, OC might be emitted directly from sources as primary particles, but secondary organic aerosols can also be formed from low vapor pressure products via atmospheric chemical reactions. At a vehicle exhaust-dominated site, the OC/EC ratios should reflect the properties of the primary source. The $PM_{2.5}$ OC/EC ratio was

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Table 1
Annual average concentrations and chemical compositions of $PM_{2.5}$ and $PM_{10\text{-}2.5}$.

	$PM_{2.5}$		$PM_{10\text{-}2.5}$		PM_{10}		$PM_{2.5}/PM_{10}$	$PM_{10\text{-}2.5}/PM_{10}$
	Average	SD[a]	Average	SD[a]	Average	SD[a]		
			μg/m³					
Mass (Teflon)	55.5	25.5	25.9	15.7	81.3	37.7	0.68	0.32
OC[b]	11.1	5.8	2.1	1.8	13.2	6.4	0.85	0.16
EC[c]	17.3	5.4	1.7	2.7	18.9	7.2	0.91	0.09
Chloride (Cl^-)	0.42	0.30	0.96	0.64	1.4	0.75	0.31	0.71
Nitrate (NO_3^-)	2.6	2.1	1.9	1.6	4.5	3.0	0.59	0.43
Sulfate (SO_4^{2-})	12.7	7.6	1.4	1.5	14.1	8.3	0.90	0.10
Soluble sodium (Na^+)	1.5	0.26	1.0	0.75	2.5	0.86	0.62	0.40
Ammonium (NH_4^+)	2.8	1.7	0.17	0.18	3.0	1.82	0.96	0.06
Soluble potassium (K^+)	0.59	0.69	0.09	0.17	0.68	0.80	0.87	0.14
Sodium (Na)	0.72	0.29	0.75	0.45	1.5	0.62	0.50	0.52
Magnesium (Mg)	0.20	0.08	0.22	0.13	0.41	0.18	0.48	0.54
Aluminum (Al)	0.30	0.19	0.69	0.55	0.96	0.73	0.31	0.71
Silicon (Si)	0.54	0.39	1.3	1.0	1.8	1.4	0.30	0.72
Phosphorus (P)	0.20	0.12	0.017	0.012	0.20	0.11	1.01	0.08
Sulfur (S)	4.9	2.9	0.39	0.27	5.2	2.8	0.94	0.07
Chlorine (Cl)	0.31	0.32	1.1	0.75	1.3	0.92	0.24	0.82
Potassium (K)	0.71	0.69	0.27	0.20	0.97	0.87	0.73	0.28
Calcium (Ca)	0.22	0.12	0.77	0.46	0.97	0.58	0.23	0.79
Titanium (Ti)	0.023	0.015	0.053	0.036	0.075	0.048	0.31	0.71
Vanadium (V)	0.018	0.018	0.0038	0.0025	0.019	0.020	0.93	0.20
Manganese (Mn)	0.031	0.023	0.020	0.013	0.050	0.030	0.63	0.40
Iron (Fe)	0.57	0.26	1.3	0.53	1.8	0.78	0.32	0.70
Nickel (Ni)	0.0058	0.0046	0.0022	0.0012	0.0066	0.0052	0.88	0.34
Copper (Cu)	0.024	0.010	0.034	0.017	0.057	0.025	0.42	0.60
Zinc (Zn)	0.30	0.25	0.060	0.052	0.35	0.27	0.86	0.17
Arsenic (As)	0.016	0.012	0.0077	0.0070	0.016	0.015	0.98	0.48
Bromine (Br)	0.014	0.013	0.0065	0.0043	0.019	0.015	0.74	0.34
Rubidium (Rb)	0.0052	0.0042	0.0047	0.0048	0.007	0.007	0.79	0.71
Strontium (Sr)	0.0048	0.0030	0.0071	0.0051	0.010	0.006	0.49	0.72
Zerconium (Zr)	0.0077	0.0043	0.0081	0.0043	0.012	0.0054	0.62	0.65
Tin (Sn)	0.029	0.019	0.013	0.011	0.034	0.025	0.85	0.38
Antimony (Sb)	0.041	0.011	0.012	0.010	0.050	0.010	0.81	0.23
Barium (Ba)	0.048	0.016	0.058	0.035	0.090	0.043	0.54	0.65
Lead (Pb)	0.061	0.059	0.016	0.017	0.069	0.067	0.88	0.23

[a] Standard deviation.
[b] OC = OC1 + OC2 + OC3 + OC4 + OP.
[c] EC = EC1 + EC2 + EC3-OP.

0.7 ± 0.3 in this study, which is consistent with the findings of Gillies, Gertler, Sagebiel, and Dippel (2001), Laschober, Limbeck, Rendl, and Puxbaum (2004), and Norbeck, Durbin, and Truex (1998). Fig. 3 shows that the peaks and valleys in the $PM_{2.5}$ OC and EC concentrations coincided over time, suggesting that vehicle exhausts were important factors. The OC/EC ratio of $PM_{10\text{-}2.5}$ (7.8 ± 14.2) was an order of magnitude higher than that of $PM_{2.5}$. Biologic aerosols and tire dust were expected to be potential sources of $PM_{10\text{-}2.5}$. $PM_{2.5}$ and $PM_{10\text{-}2.5}$ EC showed similar trends in summer, indicating impacts from local vehicle and ship emissions.

Fig. 4 shows a good correlation between $PM_{2.5}$ sulfate and ammonium, which followed a similar pattern to nitrate. Approximate 40% of $PM_{2.5}$ sulfate has been found to come from mainland China (Pathak, Yao, Lau, & Chan, 2003). $PM_{10\text{-}2.5}$ sulfate and nitrate exhibited better correlations with water-soluble sodium ($R = 0.63$ and 0.73, respectively) than with ammonium ($R = 0.31$ and 0.06, respectively) as shown in Fig. 4.

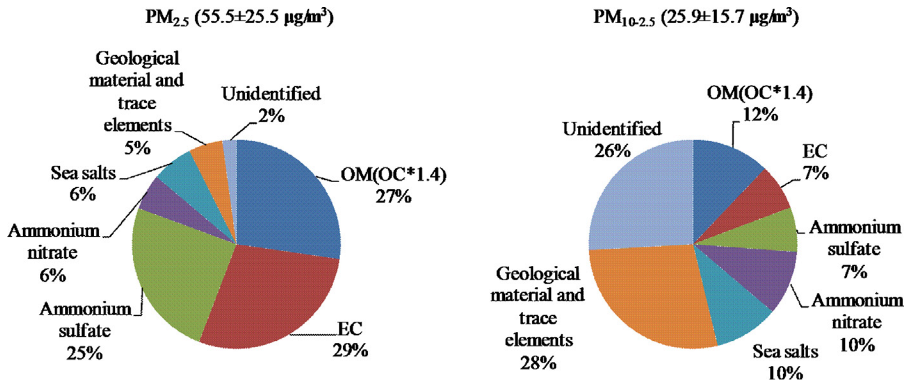

Fig. 2. Average mass closures for $PM_{2.5}$ and $PM_{10\text{-}2.5}$. The geological material was estimated as 1.89 × Al + 2.14 × Si + 1.4 × Ca + 1.43 × Fe to account for unmeasured oxides.

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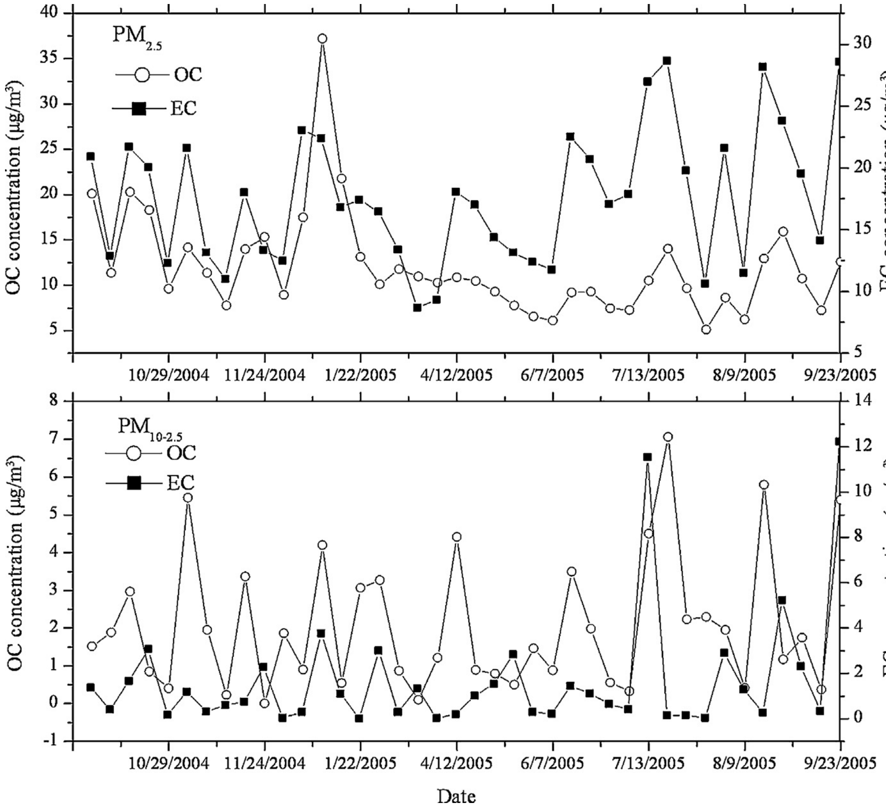

Fig. 3. Time series of OC and EC in $PM_{2.5}$ and $PM_{10\text{-}2.5}$.

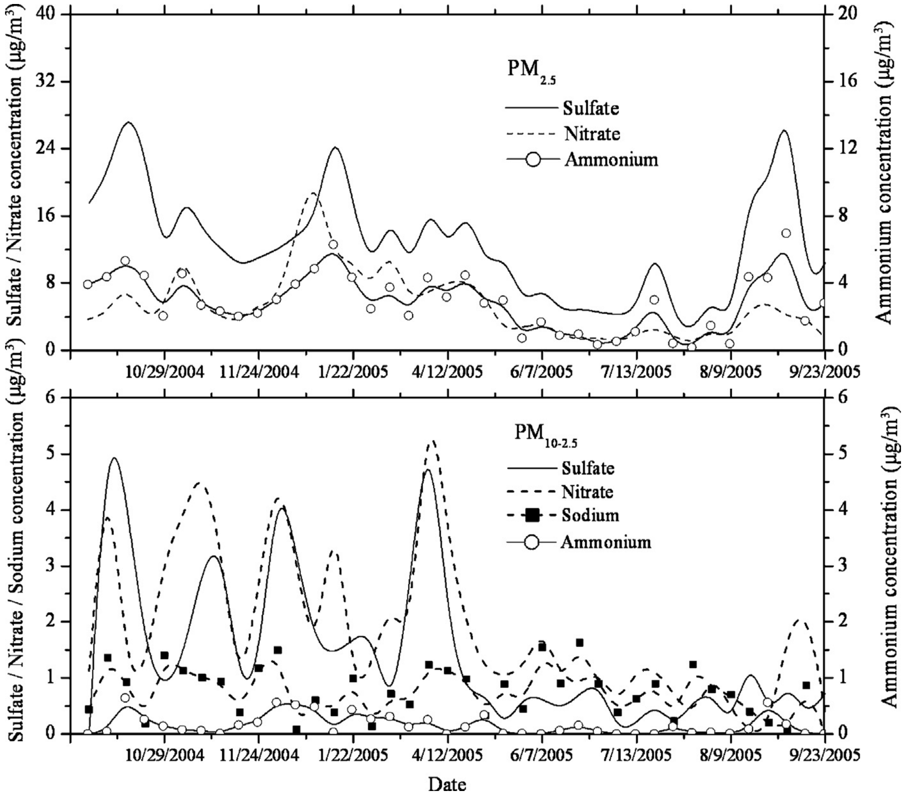

Fig. 4. Time series of water-soluble anions and cations in $PM_{2.5}$ and $PM_{10\text{-}2.5}$.

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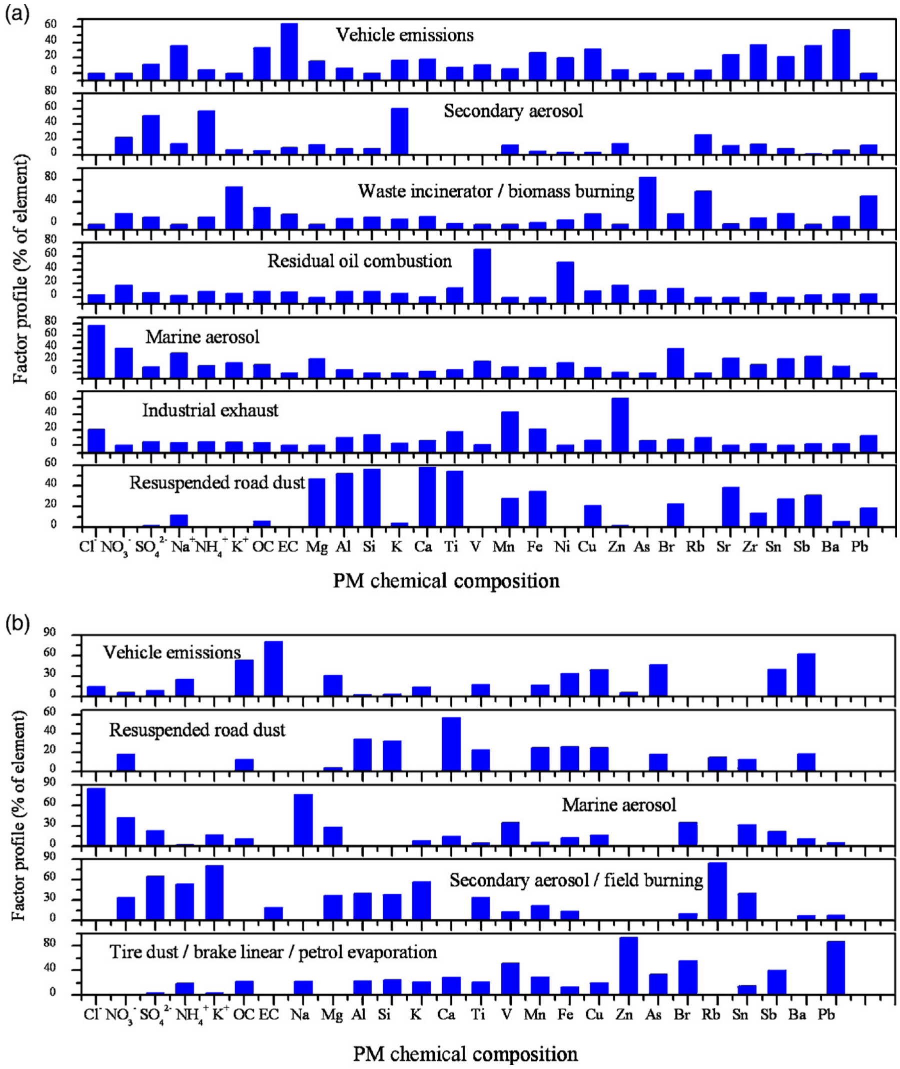

Fig. 5. Factor loadings obtained from positive matrix factorization (PMF) analysis of the chemical constituents of (a) $PM_{2.5}$ and (b) $PM_{10\text{-}2.5}$.

$PM_{2.5}$ K^+ (K) and Pb exhibited a good interspecies correlation with an R value of 0.95, which is consistent with the findings of Louie, Chow, et al. (2005) for field burning (biomass burning) and lead-fueled burning. In addition, a good relationship ($R = 0.90$) was found between Cr and Ni, which is most likely related to ship emissions from the harbor that is ~6 km from the site. Near a container terminal in Hong Kong, Yau et al. (2013) found that ship emissions accounted for ~25% of the ambient $PM_{2.5}$ mass.

$PM_{10\text{-}2.5}$ crustal elements, such as Al, Si, Ca, Ti, and Mn, showed good correlations with each other with $0.83 < R < 0.99$. These elements most likely originated from resuspended road dust. A previous study reported a strong contribution of resuspended road dust to $PM_{10\text{-}2.5}$ (Lin et al., 2005). $PM_{10\text{-}2.5}$ K and Rb ($R = 0.73$) probably resulted from burning processes as was determined for $PM_{2.5}$. As expected, $PM_{10\text{-}2.5}$ Cl and Na correlated moderately with each other with an R value of 0.78 because both elements are representative of marine aerosols (Fitzgerald, 1991).

Correlations between the different size fractions are shown in Table 2. Nearly half of the species showed good correlations between $PM_{2.5}$ and PM_{10}, but these species correlated poorly with $PM_{10\text{-}2.5}$, mainly because a majority (~70%) of PM_{10} was $PM_{2.5}$. In addition, $PM_{2.5}$ and $PM_{10\text{-}2.5}$ exhibited different properties.

3.3. $PM_{2.5}$ and $PM_{10\text{-}2.5}$ source contributions determined by the positive matrix factorization (PMF) model

The PMF model was used to determine the $PM_{2.5}$ and $PM_{10\text{-}2.5}$ source contributions. The measured concentrations and their associated uncertainties were used as model inputs. Seven sources were resolved for $PM_{2.5}$, including vehicle emissions, secondary aerosols, waste incinerator/biomass burning, residual oil combustion, marine aerosols, industrial exhaust, and resuspended road dust (Fig. 5). Vehicle exhaust was the largest contributor, accounting for ~29% of the total $PM_{2.5}$ mass as shown in Fig. 6. The PMF profile of diesel-fueled vehicles was characterized by abundant EC, followed by OC and then trace elements (e.g., Ba, Zr, Na, Sb, Cu, Fe, Sr, Sn, Ni, Ca, P, Mg, etc.). The characteristics of these profiles are identical to those from a Hong Kong tunnel study (Cheng, Lee, et al., 2010) and previous studies on vehicle emissions (Gertler et al., 2002; Norbeck et al., 1998). The second largest contributor, which accounted for ~27% of $PM_{2.5}$, was characterized by large sulfate, ammonium, nitrate, and soluble sodium contributions, suggesting it was associated with secondary inorganic aerosols. Previous studies (Cheng et al., 2006; Li et al., 2012; Li, Yu, et al., 2013; Louie, Watson, et al., 2005; Pathak, Yao, Lau, & Chan, 2003) showed the

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Table 2
Correlations (*R*) between the different size fractions.

	$PM_{2.5}$ & PM_{10}	$PM_{10-2.5}$ & PM_{10}	$PM_{2.5}$ & $PM_{10-2.5}$
Mass	**0.95**	**0.86**	0.67
OC	**0.96**	0.51	0.25
EC	**0.95**	0.76	0.51
Cl^-	0.51	**0.92**	0.14
NO_3^-	**0.84**	0.74	0.26
SO_4^{2-}	**0.99**	0.54	0.39
Na^+	0.49	**0.96**	0.23
NH_4^+	**0.99**	0.51	0.42
K^+	**0.98**	0.69	0.55
Na	0.71	**0.86**	0.30
Mg	0.71	**0.91**	0.34
Al	**0.92**	**0.99**	**0.85**
Si	**0.91**	**0.99**	**0.84**
P	**0.98**	−0.12	−0.26
S	**0.99**	0.01	−0.11
Cl	0.59	**0.95**	0.26
K	**0.99**	**0.92**	**0.88**
Ca	**0.90**	**0.99**	**0.86**
Sc	0.03	**0.85**	−0.23
Ti	**0.85**	**0.98**	0.72
V	**0.99**	0.59	0.47
Cr	0.62	**0.84**	0.09
Mn	**0.90**	0.71	0.33
Fe	**0.84**	**0.97**	0.69
Ni	−0.13	**1.00**	0.08
Cu	**0.94**	0.39	0.56
Zn	0.79	**0.94**	0.38
Ga	**0.97**	0.58	−0.25
As	0.75	0.75	0.24
Br	**0.95**	0.61	0.33
Rb	0.79	**0.90**	0.61
Sr	0.26	**0.88**	−0.19
Sn	**0.82**	0.65	0.15
Sb	0.15	0.57	−0.64
Pb	**0.97**	0.42	0.18

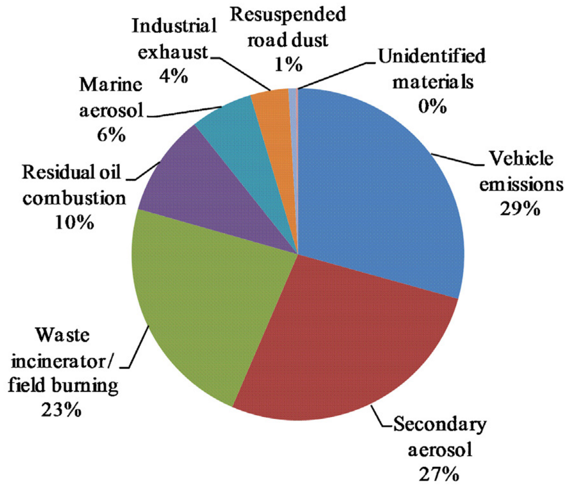
Bold values show the correlation coefficients (R) > 0.8.

effects of the long-range transport of secondary aerosols on $PM_{2.5}$ in the atmosphere of Hong Kong. Waste incinerator/field burning, which is characterized by elements such as K^+, As, Rb, and Pb, accounted for ~23% of the $PM_{2.5}$ mass (Louie, Watson, et al., 2005). Waste incinerator/field burning mainly occurs in Guang Dong Province, which is located ~40 km north of Hong Kong. The fourth source identified by PMF was residual oil combustion, which was distinguished by Ni and V and accounted for ~10% of $PM_{2.5}$ (Yu et al., 2004; Yuan et al., 2006). Spatial variations in Ni and V in Hong Kong have been associated with the location of container ports (Yu et al., 2004). Marine aerosols and industrial exhaust contributed ~6% and ~4% to the $PM_{2.5}$ mass. The remaining identified source was resuspended road dust, which was characterized by Mg, Al, Si, Ca, and Ti. The chemical profile was similar to those obtained by Ho, Lee, Chow, and Watson (2003).

Five sources were resolved for $PM_{10-2.5}$, including vehicle emissions, resuspended road dust, marine aerosols, secondary aerosols and field burning, and other vehicle-generated dust, such as tire dust, brake linear, and petrol evaporation. Four out of the five $PM_{10-2.5}$ sources were also $PM_{2.5}$ sources. The corresponding profiles for these four sources had similar indicators. Resuspended road dust, marine aerosols, vehicle emissions, and secondary aerosols/field burning contributed ~17%, ~17%, ~11%, and ~12%, respectively, to $PM_{10-2.5}$. Vehicle-generated dust sources (tire dust, brake linear, and petrol evaporation) accounted for ~20% of the total $PM_{10-2.5}$ mass. However, ~23% of $PM_{10-2.5}$ was not identified by PMF, which is consistent with the mass closure results.

(a) $PM_{2.5}$

(55.5 ± 22.5 μg/m^3)



(b) $PM_{10-2.5}$

(25.9 ± 15.7 μg/m^3)

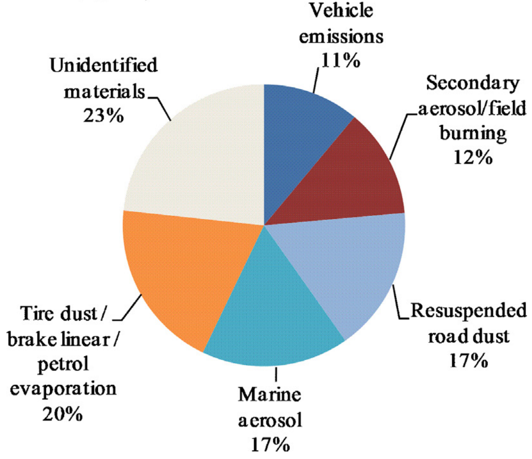


Fig. 6. Average mass concentrations and percentages of identified sources for (a) $PM_{2.5}$ and (b) $PM_{10-2.5}$.

4. Conclusions

The annual average mass concentrations were 55.5 ± 25.5 μg/m^3 for $PM_{2.5}$ and 25.9 ± 15.7 μg/m^3 for $PM_{10-2.5}$ at the PU roadside site. On average, $PM_{2.5}$ comprised ~70% of PM_{10}. The three most abundant species for $PM_{2.5}$ in descending order were EC, sulfate, and OC. $PM_{10-2.5}$ consisted of multiple components and was abundant in OC, nitrate, sulfate, Si, Fe, Cl, ammonium, and Na. The OC/EC ratios for $PM_{2.5}$ and $PM_{10-2.5}$ were 0.7 ± 0.3 and 7.8 ± 14.2, respectively, suggesting that the carbonaceous aerosols in the $PM_{2.5}$ and $PM_{10-2.5}$ size fractions originated from different sources. Low $PM_{2.5}$ OC/EC ratios (less than 1) suggested fresh vehicle exhaust was the main source. Five elements (Al, Si, Ca, Ti, and Fe) closely resembled material from the earth crust, regardless of the size range. $PM_{10-2.5}$ Cl and Na, which represented marine aerosols, correlated moderately.

The PMF model was used to determine the $PM_{2.5}$ and $PM_{10-2.5}$ source contributions. Seven sources were resolved for the $PM_{2.5}$ data, including vehicle emissions (29%), secondary aerosols (27%), waste incinerator/biomass burning (23%), residual oil combustion (10%), marine aerosols (6%), industrial exhaust (4%), and

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resuspended road dust (1%). Five sources were resolved for $PM_{10-2.5}$, including tire dust/brake linear/petrol evaporation (20%), resuspended road dust (17%), marine aerosols (17%), secondary aerosols/field burning (12%), and vehicle emissions (11%).

Acknowledgments

This project was supported by Hong Kong Polytechnic University (G-YX3L, G-YF23), the Natural Science Foundation of China (NSFC-21107084), and State Key Laboratory of Loess & Quaternary Geology (SKLLQG0804), Chinese Academy of Sciences.

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☆ This paper is adapted from the presentation at the 11th National Aerosol Conference and 10th Cross-Strait Workshop for Aerosol Science and Technology, May 16–19, Wuhan, China, as recommended by Prof. Junji Cao, the chair of the scientific committee.

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