Elemental composition of airborne aerosols at a traffic site and a suburban site in Hong Kong

Yan Cheng and Shun Cheng Lee*

Department of Civil and Structural Engineering,
Research Center for Environmental Technology and Management,
The Hong Kong Polytechnic University,
Hung Hum, Kowloon, Hong Kong, China
Fax: +86-852-2334-6389
E-mail: y.cheng@polyu.edu.hk
E-mail: ceslee@polyu.edu.hk
*Corresponding author

Junji Cao

State Key Laboratory of Loess and Quaternary Geology,
Institute of Earth Environment,
Chinese Academy of Sciences,
Xi'an, China
E-mail: cao@loess.llqg.ac.cn

Kin Fai Ho

Department of Civil and Structural Engineering,
Research Center for Environmental Technology and Management,
The Hong Kong Polytechnic University,
Hung Hum, Kowloon, Hong Kong, China
E-mail: cekfho@polyu.edu.hk

Judith C. Chow and John G. Watson

Division of Atmospheric Sciences,
Desert Research Institute,
2215 Raggio Parkway,
Reno, NV 89512, USA
E-mail: Judy.Chow@dri.edu
E-mail: John.Watson@dri.edu

Chio Hang Ao

Department of Civil and Structural Engineering,
Research Center for Environmental Technology and Management,
The Hong Kong Polytechnic University,
Hung Hum, Kowloon, Hong Kong, China
E-mail: crispinao@gmail.com

Abstract: Forty elemental concentrations in different size particles were measured at a roadside site and a suburban site of Hong Kong using a ten-stage MOUDI impactor (Mode 110). Some elements (Cr, Fe, Co, Cu, As and Ba) have clear spatial variability, with ~100% higher concentrations in roadside atmosphere than in suburban atmosphere, showing obviously vehicle-related sources for those elements. However, it is contrary for three typical sea salt elements (Na, Cl and Br), showing more than 100% higher concentrations in suburban environment than in roadside environment, which is due to the influence of marine aerosols. The size distributions of elements are generally similar between roadside and suburban site. The elements Na, Mg, Al, Si, Cl, Ca, Ti, Mn and Fe mainly present in coarse mode (10 μm > diameter > 1 μm). The mass concentrations of K, V and Ni are distributed in both fine (diameter > 1 μm) and coarse mode. The size distributions of S, Pb, As, Se, Zn, Cu, Cd and Ba show a single peak at around 1 μm or less. The contributions of mineral dust to Total Suspended Particles (TSP) are similar for the traffic and suburban site, ~13.8% and ~14.2%, respectively. Sea salts account for ~1.6% of TSP for the traffic site, and ~6.0% for the suburban site.

Keywords: element; size distribution; mineral dust; sea salts; Hong Kong.

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Biographical notes: Yan Cheng is a PhD student at the Department of Civil and Structural Engineering, Hong Kong Polytechnic University. Her major is Environmental Sciences and recent research interests are urban air pollution. She takes part in many researches such as Research Grants Council of Hong Kong and Area of Strategic Development on Atmospheric and Urban Air Pollution funded by The Hong Kong Polytechnic University.

Shun Cheng Lee is currently a Professor and Air Laboratory in-charge in the Department of Civil and Structural Engineering at the Hong Kong Polytechnic University (HKPU). He received his PhD from Berkeley in 1994. He has over 12 years of experience in conducting air quality studies and performing statistical data analysis. He has been principal investigator on indoor and outdoor air quality research studies. He has published more than 60 peer-reviewed papers.

Junji Cao is an Associate Professor and Executive Director at the Aerosol and Environment Division, Institute of Earth Environment, Chinese Academy of Sciences (CAS). He holds a PhD in Quaternary Environment. His recent research interests are carbonaceous aerosol and atmospheric chemistry, atmospheric particulate pollution and paleo-aerosol cycle and past global change. He has published more than 60 papers including 30 papers in international journals. He is a Member of editorial review board of *Journal Aerosol and Air Quality Research*.

Kin Fai Ho is currently the Post-Doctoral fellow of the Department of Civil and Structural Engineering at the Hong Kong Polytechnic University (HKPU). His recent research interests include carbonaceous aerosols, in particular the physical and chemical characteristic of organic and elemental carbon in different rural and urban areas (sources and ambient studies) as well as atmospheric chemistry studies and visibility deterioration in Pearl River Delta Region.

Judith C. Chow is a Research Professor in the Atmospheric Sciences Division (AS) at the Desert Research Institute (DRI), University and Community College System of Nevada. She has over 22 years of experience in conducting air quality studies and performing statistical data analysis. She directs DRI's Environmental Analysis Facility where she supervises filter processing and chemical operations and develops cost-effective, yet accurate, methods for aerosol sampling and analysis. She is the principal author or co-author of more than 100 peer-reviewed publications and more than 150 technical reports. She has been principal investigator or a major collaborator in more than 40 large air quality studies.

John G. Watson is a Research Professor in the Atmospheric Sciences Division (AS) at the Desert Research Institute (DRI), University and Community College System of Nevada. He has over 25 years of experience in the environmental sciences, including the conduct and management of major air quality studies designed to evaluate and solve specific pollution problems. His research includes the development and evaluation of measurement process, receptor models for source apportionment, causes and amelioration of urban and regional haze, fugitive dust and motor vehicle emissions and the effects of measurement uncertainty on model results. He has published more than 200 peer-reviewed papers and book chapters.

Chio Hang Ao is an Assistant Environmental Consultant in Hong Kong. He obtained his PhD at The Hong Kong Polytechnic University. His research interests include bioaerosol disinfection and indoor air pollution control by nano-photocatalytic TiO_2 technology. He has filed two patents for such technology development. He was also the project coordinator for a consultancy project, whose aim is to evaluate the pollutants emitted from different cuisines (VOCs, carbonyls, PAHs, $PM_{2.5}$, OC/EC) and its correlation between different cooking styles.

1 Introduction

In the last few decades, there has been a growing concern on environmental contamination by trace elements. It is reported that the remote and polar areas contain a large number of trace metals that are believed to be transported from continent (Rosman et al., 1994; Wiersma and Davidson, 1986). For human health, Nriagu and Pacyna (1988) find that the toxic metals have accumulated in the human food chain and Berggren et al. (1990) proved that the concentrations of trace metals have also exceeded critical thresholds, which would cause toxic effects within the terrestrial ecosystems.

Trace metals are found in almost all size fractions of atmospheric aerosols. Particle size is an extremely important parameter to consider when assessing the potential toxicity and sources of metals present in urban aerosols (Natusch and Wallace, 1974). Coarse inhalable particles may be deposited in the upper respiratory tract whereas fine particles travel deeper into the lungs. The finest particles can reach alveolar regions (Voutsa and Samara, 2002). Ultrafines can more efficiently transfer the catalytic metals on the surface of irritant particles to the lungs than can an equal mass of larger particles (Hughes et al., 1998).

The origin of trace elements includes both anthropogenic and natural sources, of which manmade sources significantly exceed natural contribution for some elements (Pacyna, 1986a). Human activities, such as energy generation, industrial metal production and vehicular traffic, can result in a dramatic increase in trace element emissions to environment. The most common natural sources are windblown dusts, volcanic eruption, airborne sea salts and biological mobilisation.

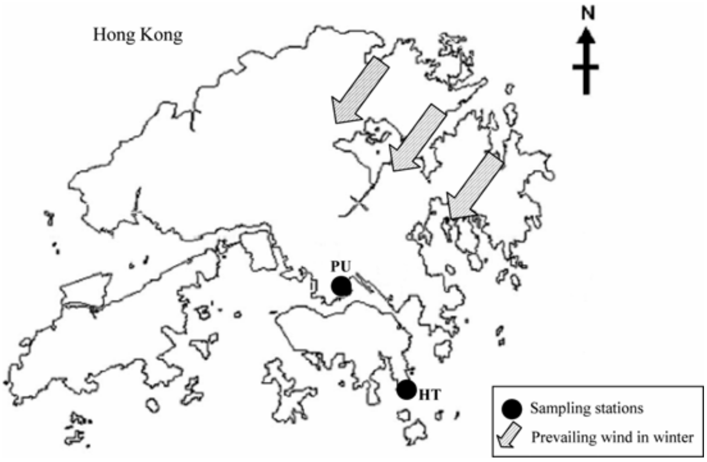
In an urban area, vehicle-related emissions contribute significantly to ambient atmosphere. Emission inventories in Hong Kong show that PM_{10} emitted from vehicles accounts for about 30% of total respirable suspended particulates in ambient atmosphere (http://www.epd.gov.hk). Therefore, it is necessary to understand the levels and size distributions of trace elements in particles, especially for toxic elements due to vehicle emissions. In fact, some toxic elements (Ba, Ni and Pb) are typically present in vehicle exhausts (Lough et al., 2005; Pacyna, 1986b). However, very limited results for elemental size distributions are available in Hong Kong because most of the previous studies (Ho et al., 2003a; Louie et al., 2005) only focused on elemental concentrations in $PM_{2.5}$ or PM_{10}. Here we report the concentrations of elements and their size distributions monitored at a traffic site and make a comparison with a suburban site, to improve our understanding of the current aerosol chemical speciation information from major sources in Hong Kong area.

2 Sampling and experimental procedures

2.1 Sampling site

From November 2003 to February 2004, three sets of size-selective samples were collected in the campus of the Hong Kong Polytechnic University (PU, Figure 1), a typical roadside location in urban Hong Kong impacted mostly by primary vehicular emissions. The sampling site in PU is situated at ground level, about 1 m away from the main traffic road. The traffic flow on the road is extremely high, with about 119,759 per day (the 2003 annual traffic census).

Figure 1 Sampling stations in Hong Kong



For comparison, additional three sample sets were obtained at a suburban site, Hok Tsui (HT, Figure 1) site and the sampling period was from February to March 2004. HT site is located in the southern most edge of Hong Kong Island with 240° of ocean view from northeast to southwest, about 60 m above sea level. Because the prevailing surface wind in HT is from southeastern China in winter, the samples are expected to be influenced by the polluted air masses of long-range transport from the mainland China (Wang et al., 1997, 2003a,b).

2.2 Sample collection

A ten-stage MOUDI (MOUDI 110) with a flow of 30 l min^{-1} was used to collect samples for 48-hr periods at PU site, while 96–120 hr at HT site due to low particulate concentrations. Particles are classified by the ten-stage MOUDI in the following aerodynamic particle diameter, 0.056, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 2.5, 5.6 and 10 μm. Additional two stages including inlet stage (>18 μm) and backup filter were used here and the assumed the highest and the lowest 'cut size' was 100 μm and 10 nm, respectively. Teflon membranes (Pall Life Science, Michigan, USA) with diameter of 47 mm were used to collect particles in the upper eleven stages. For particles with diameters less than 0.056 μm were collected on backup filters with 37 mm Teflon membrane filters. The flow rates were checked in the field before and after each run using a calibrated flow meter. The differences of flow rates were within ~5%. At the end of each sampling interval, all loaded filters were placed into plastic petri dishes and then all petri dishes were put into a sealed plastic bag before quickly transported to a laboratory. The samples were stored in a refrigerator to prevent the evaporation of volatile components and then sent to DRI (packed in blue ice) for chemical analyses. During the sampling periods in our studies, the Relative Humidity (RH) was high with RH values of 60–80%, thus the effect of particle bounce was neglected in sampling, as previously demonstrated by Stein et al. (1994) and Vasiliou et al. (1999).

2.3 Gravimetric analysis

To determine particle mass by gravimetric analysis, all filters were pre- and post-weighed at least twice and the net weights were obtained by subtracting the initial weights from the final weights. Before weighing, all filters were exposed for a minimum of 24 hr at the equilibration condition with temperature of 20–23°C and RH of 30–40%. The filters were weighted using a Microbalance (Model MC5, Sartorius AG, Goettingen, Germany) with the sensitivity of ±1 μg in the range of 0–250 mg. The net weights were divided by the total sampling volume with a consideration of the temperature and pressure during sampling periods, thus the mass concentrations can be obtained with high resolution. The filters are handled only with tweezers cleaned by dry KimWipes (Kimberly-Clark Corporation, USA) to reduce the possibility of contamination.

2.4 Elemental analysis

Forty elements on Teflon filters were determined in Desert Research Institute, USA, with the energy dispersive X-Ray Fluorescence (XRF) analytical method (Watson et al., 1999), including Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Au, Hg, Tl, Pb and U. A multielement thin film standard from μ Matter, Inc. was used to be reference materials during analysis. Replicated analyses were performed for ~10% of all samples. The differences for replicated samples were within ~5%. Also, three blank samples were analysed to correct the concentrations of individual specified species.

XRF spectroscopy is based on the measurement of X-ray energy produced by the ejection of an inner shell electron from an atom in the sample. The X-ray is specific for different atoms and the intensity of the X-ray is proportional to the number of atoms in the sample. Therefore, the concentration of elements can be quantitatively determined through a comparison with the known standards.

3 Results and discussion

3.1 Mass and elemental concentrations

Arithmetic means and standard deviations of total particle masses and 40 elements in aerosol particles are summarised in Table 1. The average mass concentration of Total Suspended Particles (TSP) is 70.7 ± 5.3 $\mu g\ m^{-3}$ and 50.2 ± 8.0 $\mu g\ m^{-3}$ for PU and HT site, respectively. The three elements with the highest average concentrations in the TSP samples at PU site are S (3001.2 ± 826.2 $ng\ m^{-3}$), Si (2037.6 ± 1355.0 $ng\ m^{-3}$) and Fe (1599.0 ± 659.8 $ng\ m^{-3}$), accounting for 96.1% of the total measured elements. While in HT site, Cl (4969.9 ± 885.5 $ng\ m^{-3}$) is found to be the most abundant constituent, followed by S (2348.3 ± 1112.7 $ng\ m^{-3}$) and Si (2155.2 ± 463.2 $ng\ m^{-3}$) and these three elements contribute to 98.0% of the total measured elements in HT site. Both of S and Si are abundant in PU roadside and HT suburban atmosphere, suggesting the ubiquitous distribution of the two elements in atmosphere. Nearly all of sulphur elements present as sulphate in the atmosphere of Hong Kong (Chow et al., 2002). The major sources of S in atmosphere are SO_2 and H_2SO_4 from combustion of sulphur-containing fuel and the contribution of S to the earth's crust is minor (Taylor and McLennan, 1995). Pathak et al. (2003) claimed that a considerable number of sulphate (~40%) in ambient atmosphere of Hong Kong is from Asian continental air masses, which may be the reason for even distribution of S. HKEPD (Environmental Protection Department in Hong Kong) has recognised that the regional pollutants resulted in deteriorating air quality within the Hong Kong area. Some effective control strategies have been developed through a joint study of air quality in the Pearl River Delta Region, launched by the HKEPD and the Guang Dong Province Environmental Protection Bureau (GDEPB) in 2002. Therefore low levels of regional pollutants, like S, are expected in future. Si is generally considered being from nearby resuspended geological material, although trace Si has also been observed from other sources, such as coal fly ash (Husain, 1986), ferroalloy manufacture (Pacyna, 1986a) and vehicle emissions (Watson and Chow, 2001). The Cl abundance obtained in HT site is reasonable considering its location of marine surroundings.

It is very useful to use enrichment factor to examine the origin of elements. Based upon the assumption that all detected Al in each size range originate from resuspended soil material, the earth crust is considered the dominant source of the elements that show EF values close to one using Al as a reference element ($EF_{crust} = (X/Al)_{air}/(X/Al)_{crust}$). Elements with EF_{crust} value larger than about five are called enriched elements and have some sources other than crustal weathering, which may be anthropogenic. Based on the results of EF analyses (Table 1), eight elements (Na, Mg, Al, Si, P, Ti, Sr and Zr) at PU roadside station closely resemble those of material from the earth crust (Taylor and

McLennan, 1995). On the contrary, only four crustal elements (Mg, Al, Si and Ti) are observed in HT site.

For the remaining elements with the EF$_{crust}$ values larger than five at both of PU and HT site, they are significantly influenced by anthropogenic sources. The levels of most non-crustal and anthropogenic elements have clear spatial variability of high concentrations in roadside atmosphere and low concentrations in suburban atmosphere, excluding several typical sea salt elements (Na, Cl and Br) and elements in concentrations below the minimum detectable limit (Ga, Mo, In, Sb and Au). The concentrations of some elements (Cr, Fe, Co, Cu, As and Ba) in PU site is about 100% higher than those in the HT site, and it clearly illustrates that these elements are associated with anthropogenic sources, most likely the vehicle-related emissions. Several studies have previously shown the enrichment of similar elements in vehicular engine exhausts (Wang et al., 2003a,b; Yeung et al., 2003). Elements Cl and Na, two typical elements in sea salt aerosols, are more than 100% higher at HT site than those at PU site, reflecting the impact of marine aerosols around HT sampling station.

Table 1 Concentrations of elements for TSP as well as enrichment factors at PU and HT

Mass	*Unit*	*MDL*[a]	*PU*			*HT*		
			TSP	*SD*[b]	*EFcrust*[c]	*TSP*	*SD*[b]	*EFcrust*[c]
Mass	μg m^{-3}		70.7	5.3		53.2	8	
Na	ng m^{-3}	33.1	571.5	246.9	4.6	1073.2	112.2	11.6
Mg	ng m^{-3}	12	104.7	26.1	1.8	133.1	31.8	3.1
Al	ng m^{-3}	4.8	342.7	214.8	1	256.3	51.4	1
Si	ng m^{-3}	3	2037.6	1355	1.6	2155.2	463.2	2.2
P	ng m^{-3}	2.7	6.6	2.3	2.2	0	0	0
S	ng m^{-3}	2.4	3001.2	826.2	No data	2348.3	1112.7	No data
Cl	ng m^{-3}	4.8	1102.5	562.2	No data	4969.9	885.5	No data
K	ng m^{-3}	2.9	941	679.6	7.9	585.2	19.2	6.6
Ca	ng m^{-3}	2.2	1007.9	632.4	7.9	711.6	141.8	7.4
Ti	ng m^{-3}	1.4	45.4	38.1	3.6	45.8	10.5	4.8
V	ng m^{-3}	1.2	7.6	1	29.7	9.8	4.3	51.2
Cr	ng m^{-3}	0.9	8.8	2.5	59	3.2	1	28.7
Mn	ng m^{-3}	0.8	36.4	27.5	14.2	25.3	8.4	13.2
Fe	ng m^{-3}	0.7	1599	659.8	10.7	637.2	221.8	5.7
Co	ng m^{-3}	0.4	8.4	4	197.1	3.8	2.3	119.2
Ni	ng m^{-3}	0.4	4.7	0.8	55.1	5.1	0.7	80
Cu	ng m^{-3}	0.5	55.5	14	520.8	7.8	1.6	97.9
Zn	ng m^{-3}	0.5	296.2	175.5	978.7	183.9	41.6	812.5
Ga	ng m^{-3}	0.9	0.7	0.4	9.7	0.2	0.2	3.7
As	ng m^{-3}	0.8	8.7	8.1	1360.7	3.9	0.9	815.6
Se	ng m^{-3}	0.6	2.7	1.6	12.7	2.2	0.2	13.8
Br	ng m^{-3}	0.5	8.9	2.1	No data	34.1	10.6	No data
Rb	ng m^{-3}	0.5	6.9	5.4	14.5	4.8	1.6	13.4
Sr	ng m^{-3}	0.5	6.5	3.6	4.4	11.1	3.8	9.9
Y	ng m^{-3}	0.6	1.2	0.4	12.8	1	0.4	14.3

Table 1 Concentrations of elements for TSP as well as enrichment factors at PU and HT (continued)

Mass	*Unit*	*MDL*[a]	*PU*			*HT*		
			TSP	*SD*[b]	*EFcrust*[c]	*TSP*	*SD*[b]	*EFcrust*[c]
Zr	ng m^{-3}	0.8	6.1	3	7.5	3.4	1.4	5.6
Mo	ng m^{-3}	1.3	2.8	0.5	437.9	0.7	0.3	146.4
Pd	ng m^{-3}	5.3	0.2	0.1	93843	0.1	0	62739
Ag	ng m^{-3}	5.8	1.2	0.3	5645.5	0.7	0.3	4391.7
Cd	ng m^{-3}	5.8	2.4	2.3	5745.5	1.3	0	4161.3
In	ng m^{-3}	6.2	1.6	0.3	7507.4	0.3	0.3	1882.2
Sn	ng m^{-3}	8.1	36.5	15.4	1556.9	11.5	1.9	655.9
Sb	ng m^{-3}	8.6	17	4.5	19941.6	3.4	0.6	5332.8
Ba	ng m^{-3}	24.9	76.7	25.8	32.7	21.5	3.9	12.3
La	ng m^{-3}	29.7	10	9.9	78.2	5.1	0.8	53.3
Au	ng m^{-3}	1.5	0.5	0.4	65168.8	0	0	0
Hg	ng m^{-3}	1.2	0.4	0.3	No data	0.3	0.1	No data
Tl	ng m^{-3}	1.2	1.1	1.3	344.1	1.1	0.2	460.1
Pb	ng m^{-3}	1.4	91.4	73.7	1072.2	61.5	12.1	964.6
Ur	ng m^{-3}	1.1	0.3	0.2	25.1	0.2	0.1	22.4

[a] Minimum detectable limit (MDL) is the concentration at which instrument response equals three times the standard deviation of the response to a known concentration of zero.

[b] Standard Deviation.

[C] Enrichment factor (EFcrust) = (X/Al)air/(X/Al)crust.

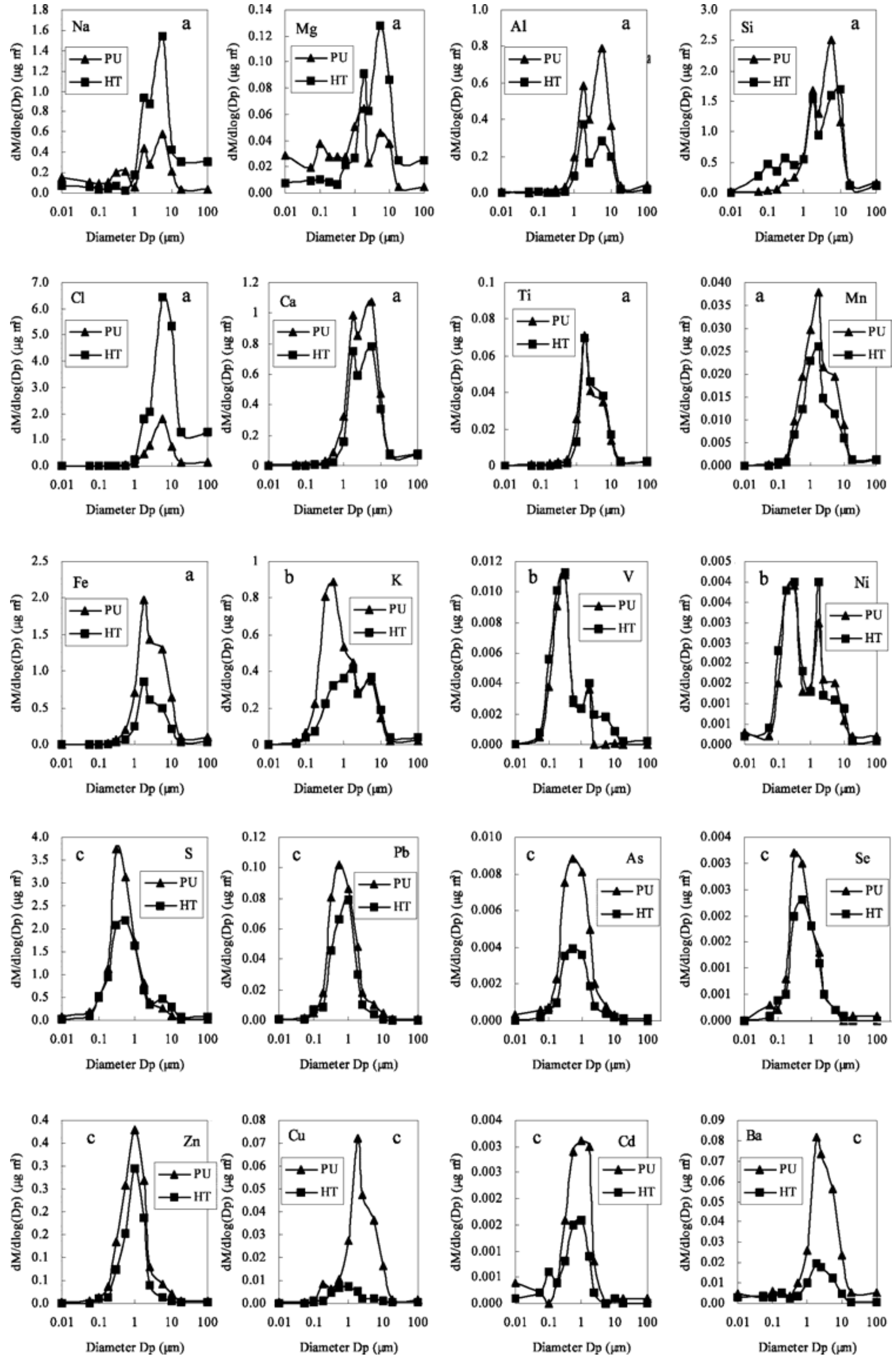
The situations of heavy metals in atmospheric aerosols are of particular interest to toxicologists because of their toxicity, ubiquitousness, their catalytic function on oxidative reactions and the fact that it is not easy to be chemically transformed or destroyed. Several elements are classified by the US EPA as air toxics, such as Cr, As, Cd and Pb. The concentrations of Cd and Pb obtained at PU roadside station is 2.4 ± 2.3 ng m^{-3} and 91.4 ± 73.7 ng m^{-3}, respectively, which is higher than previously reported ambient levels in 2002 (1.43 and 56 ng m^{-3} for Cd and Pb, respectively, 2002 air quality report of EPD, http://www.epd.gov.hk) by a factor of 1–2, implying the presence of anthropogenic sources for Cd and Pb at PU roadside station, namely the fuel combustion from vehicles. The levels of Cd (1.3 ± 0.0 ng m^{-3}) and Pb (61.5 ± 12.1 ng m^{-3}) observed at HT site are quite close to ambient annual levels, suggesting the low contributions from vehicles for the two elements. The low levels of Pb are not surprised because leaded petrol has been banned in Hong Kong since 1 April 1999.

3.2 Size distributions of major elements

As illustrated in Figure 2, the size distributions of major elements can be divided into three groups:

1 elements that mainly present in coarse mode
2 elements that distribute in both of fine and coarse mode and
3 elements with a single peak at around 1 μm or less.

Figure 2 Size distribution of major elements measured in PU roadside and HT suburban environment. a, b and c represents Groups 1, 2 and 3, respectively.



3.2.1 Group 1 (Na, Mg, Al, Si, Cl, Ca, Ti, Mn and Fe)

The size distributions of Mg, Al, Si and Ca are quite similar between PU and HT site, showing dual modes peaking at 1.8 and 6.8 μm, respectively. The dominant mass is found in the coarse mode, amounting to over 70%. The size distributions of Ti, Mn and Fe are also bimodal at both of PU and HT site, but the coarse mode contributes much less to mass than the fine mode. Patterson and Gillette (1977) claimed that the typical soil-derived elements, including above seven elements (Mg, Al, Si, Ca, Ti, Mn and Fe), generally present in diameter of 1–10 μm and possibly result from the breaking up of the aggregates by a process of sandblasting and subsequent injection of the disaggregated material into the atmosphere. In this study, the high EF values for element Ca, Mn and Fe seem to imply that soil material is not the only source. This maybe attributed to the contamination of roadway soil with previously deposited particles emitted from motor vehicles. The elements Fe and Mn were found to be the main metal pollutants emitted by vehicles in a previous study (Monaci et al., 2000). Mn was also found to associate with brake dust (Adachi and Tainosho, 2004). Ho et al. (2003b) and Yeung et al. (2003) reported higher Ca levels in road dust than urban soils in Hong Kong. It is noticed that there is a trimodal distribution for Mg at PU station. Besides two similar coarse modes to other soil-derived elements, the third mode exists in accumulation size fraction centered at 0.1 μm. This specific component is believed to be from vehicle exhausts because particles in accumulation size range are generally related to anthropogenic combustion.

Cl has one predominant coarse mode, peaking at about 6 μm in the atmosphere of roadside and suburban sites, which is consistent with sea salts aerosol. The size distribution of Na at HT station is similar to other typical crustal elements, such as Mg, Al and Si, while the concentration is quite high exceeding the levels of pure crustal Na. This demonstrates that a non-crustal source, most likely sea salts, also contributes to Na. The third peak of Na in accumulation size range is only observed at PU roadside site, which is related to fuel combustion from the vehicles.

3.2.2 Group 2 (K, V and Ni)

The size distribution of K measured at PU shows a trimodal size distribution, peaking at 0.4, 1.8 and 6.8 μm, respectively and the mass distributes relatively high in accumulation size range (over 50%) and low in two coarser size ranges (equally 20% for each). The first two peaks obtained in particles are quite similar to the size distribution of K measured from vehicle exhausts with dilution chamber (Kleeman et al., 2000), but both of the centres slightly shift to larger size fractions, possibly due to the particle condensation or coagulation during the dilution process after emitting from vehicles. Measured K in coarse mode is likely to be related to crustal material since crustal elements are the most abundant metals in the coarse mode. Thus the source of K observed at PU roadside station is a mixture of fuel combustion and road dust resuspension. The first peak of K observed at PU was obscure at HT station, indicating less anthropogenic combustion.

In this study, V and Ni show similar trimodal distributions of element K at PU roadside station and HT suburban station, with the dominant mass in accumulation mode. The similar size distributions at two locations for V and Ni suggest that they are from similar sources. In a previous study conducted by Yu et al. (2004), element V and Ni were found to mainly originate from ship emissions and the levels depend on the distance from (or relative location to) the city's container port (Yu et al., 2004).

3.2.3 Group 3 (S, Pb, As, Se, Zn, Cu, Cd and Ba)

All elements in the third group are found in concentrations below 100 ng m^{-3}, except for S. S is one of abundant elements in atmosphere of Hong Kong and has a dominant accumulation mode for both roadside (PU) and suburban environment (HT), centring at about 0.4 μm. This result agrees to a previous finding that a significant mass fraction of sulphate (SO_4^{2}) exists in the accumulation mode in both of continental and marine atmosphere (Whitby, 1978). Size distributions of Pb, As and Se show that the concentration is mostly dominated in the submicron size with a centre at 0.6–1 μm in PU and HT, with higher concentrations at PU and lower concentrations at HT.

Higher levels of Zn have been measured in street dust than in other urban area of Hong Kong (Ho et al., 2003b). In the present study, element Zn in roadside atmosphere is about two times higher than in suburban atmosphere. The Zn in roadside and suburban environments in this study is exclusively manmade, according to the EF analysis. It is known that zinc oxide is added as a vulcanisation agent in the tyre tread. The diameter of particulate ZnO was reported to be about 1 μm or less (Adachi and Tainosho, 2004), which agrees well with the size distribution of Zn observed in this study (see Figure 2). This further proves that the tyre tread is the dominated source for Zn observed in this study. The size distributions of Cu, and Ba are similar to Zn, with single mode peaking at about 1 μm. They were found to originate from break dust (Adachi and Tainosho, 2004).

4 Major natural sources: mineral dust and sea salts

In Hong Kong, vehicle exhausts are the major anthropogenic sources for particles (http://www.epd.gov.hk), and mineral dust and sea salts are the dominating natural sources. To evaluate the contribution from natural sources, the mineral dust and sea salts were calculated from elements measured at both of PU and HT. The mineral dust was calculated as introduced by Solomon et al. (1989), which is the sum of the aluminium, silicon, calcium and iron oxides (i.e. [1.89 × aluminium] + [2.14 × silicon] + [1.4 × calcium] + [1.43 × iron]). The non-crustal Na (the difference between total and crustal Na) was used to estimate the amount of sea salts in the total aerosols. Crustal Na is the product of the Al concentration and the crustal Na/Al ratio (0.36, Taylor and McLennan, 1995). Based on the assumption of that non-crustal Na is the same as Na in sea salts, sea salts concentration can be determined according to the proportion of Na in sea salts (i.e. sea salts aerosol = 3.256 × Na, by weight, Prospero, 2002).

The reconstructed concentrations of mineral dust and sea salts at HT is 7.6 and 3.2 μg m^{-3}, respectively, and 9.8 and 1.1 μg m^{-3} at PU station. Thus the amounts of mineral dust and sea salts account for about 14.2% and 6.0% of the TSP at HT, and 13.8% and 1.6% for PU. The residual 79.8% of mass in HT and 84.6% at PU are assumed to be anthropogenic.

5 Conclusion

In this study, particle concentrations and the size distributions of elements were measured in a roadside (PU) and in a suburban environment (HT) of Hong Kong from November 2003 to March 2004. S (3001.2 ± 826.2 ng m^{-3}), Si (2037.6 ± 1355.0 ng m^{-3}) and Fe (1599.0 ± 659.8 ng m^{-3}) are the most abundant elements presented at PU, accounting for 96.1% of total measured elements. While Cl (4969.9 ± 885.5 ng m^{-3}), S (2348.3 ± 1112.7 ng m^{-3}) and Si (2155.2 ± 463.2 ng m^{-3}) are found to be the most abundant constituents at HT, accounting for 98.0% to the total measured elements.

Eight elements (Na, Mg, Al, Si, P, Ti, Sr and Zr) at PU roadside station closely resemble those of material from the earth crust (Taylor and McLennan, 1995). However, only four crustal elements (Mg, Al, Si and Ti) are observed in HT site. The levels of most non-crustal and anthropogenic elements have clear spatial variability of high concentrations in roadside atmosphere and low concentrations in suburban atmosphere, excluding several typical sea salt elements (Na, Cl and Br) and elements in concentrations below the minimum detectable limit (Ga, Mo, In, Sb and Au). The concentrations of some elements (Cr, Fe, Co, Cu, As and Ba) in PU site is about 100% higher than those in the HT site, and it clearly illustrates that these elements are associated with anthropogenic sources, most likely the vehicle-related emissions. Elements Cl and Na, two typical elements in sea salt aerosols, are more than 100% higher at HT site than those at PU site, which is due to the influence of marine environment around HT sampling station.

Based on the size distributions of elements, three kinds of elements are found at both PU and HT. The first group is elements that are mainly present in coarse modes, originating from soil-derived materials (Mg, Al, Si, Ca and Ti) and sea salts (Na and Cl). The second group includes elements distributed in not only fine mode but also coarse mode (K, V and Ni). The sources of K at roadside station are found to be a mixture of fuel combustion from vehicles and suspended dust. V and Ni have the same anthropogenic source of ship emissions for both of PU and HT. Elements in the third group have a single peak at around 1.0 μm or less (S, Pb, As, Se, Zn, Cu, Cd and Ba). They are related to vehicles, such as fuel combustion, tyre tread or brake dust.

Mineral dust and sea salts are major natural sources for particles in PU and HT atmosphere, accounting for about 14.2% and 6.0% of the TSP at HT and around 13.8% and 1.6% at PU, respectively. The residual of mass in HT and PU are assumed to originate from anthropogenic sources.

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