Source apportionment of PM$_{2.5}$ in urban area of Hong Kong

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Abstract

A monitoring program for PM$_{2.5}$ had been performed at two urban monitoring stations in Hong Kong from November 2000 to February 2001 and June 2001 to August 2001. PM$_{2.5}$ samples were collected once every 6 days at PolyU and KT stations with the sampling duration of 24-h. A sum of 25 chemical species in PM$_{2.5}$ were determined and selected for receptor models. Enrichment factors relative to earth crust abundances were evaluated and it was noted that most crustal elements including Al, Ti, Mg, Ca and K have small enrichment factors. Correlation and multivariate analysis technique, such as principal components analysis (PCA)/absolute principal components analysis (APCA) and cluster analysis (CA) are used for source apportionment to identify the possible sources of PM$_{2.5}$ and to determine their contribution. Six factors at each site were isolated by using PCA/APCA and cluster analysis. Similar sources (crustal matter, automobile emission, diesel emission, secondary aerosols, tire wear, and non-ferrous smelter) are identified by the PCA/APCA and cluster analysis.
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1. Introduction

Receptor models provide a theoretical and mathematical framework for quantifying source contributions. They interpret measurement of physical and chemical properties taken at different times and places to infer the possible or probable sources and to quantify the contributions from those sources [1]. The purpose of a receptor source apportionment model is to estimate the contributions of specific source types to pollutant levels in the atmosphere at a sampling (or receptor) site. The contributions of each source are distinguished through differences in their physical and chemical properties. Computer-generated source apportionment results must be interpreted by those with knowledge of the site and the associated potential sources.

Principal component analysis (PCA) is one of the oldest and most widely used multivariate statistical techniques in the atmospheric sciences [2]. Usually the data for atmospheric aerosols exhibit many large correlations among parameters and PCA results in a much more compact representation of their varia-

tions. By using PCA, Saucy (1991) was able to identify three major sources that contributed to the atmospheric aerosol (near Phoenix, Arizona), namely crustal material, copper smelters and marine air [3]. Fung and Wong (1995) sampled total suspended particulates (TSP) in the western part of the New Territories in Hong Kong between 1986 and 1987 and analyzed various trace metals (e.g. Se, As, Sr, V, etc) as markers [4]. Then PCA was applied to identify the sources and the mass contributions of each source obtained. In many recent source apportionment studies, quantitative aerosol source apportionment was performed using absolute principal component analysis [5–8]. The APCA model can determine: (1) the number of relevant source types influencing the receptor site, (2) the source profiles of these sources in absolute numbers and, finally (3) the impact that each source type has on the concentration levels of the measured air pollutants at the receptor site [5].

The cluster analysis is another effective multivariate statistical method. Although cluster analysis is a potentially useful technique for grouping samples, its application to atmospheric studies has not been attempted broadly. One of the reasons might lie in the difficult interpretation related to the dendrograms. Environmental variables may force unclear sample groups such that the dendrogram is difficult to interpret. In

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spite of this, it is considered that cluster analysis should be performed, at the very least, to confirm the sample score groups [9]. Saucy (1991) also coupled cluster analysis with principal component analysis to examine compositions and time-dependent concentrations of aerosol particles; they revealed 15 chemically distinct particle types from the samples [3]. Most source identification/apportionment applications have been based on inorganic aerosol components, primarily trace elements often combined with ionic components [10,11]. Moreover, previous work on receptor modeling study of aerosols in East Asia is rather limited [4,12–15]. The objective of this study was to isolate, identify and quantify possible sources that contributed to fine particulate matter (PM$_{2.5}$) in urban area of Hong Kong.

2. Methodology

2.1. Sampling sites

Two sampling sites including The Hong Kong Polytechnic University (PolyU campus) and Kwun Tong (KT) were selected for PM$_{2.5}$ monitoring. The field descriptions were given as follows (Fig. 1).

PolyU campus: It situated at about 6 m above ground level and about 8 m away from the main traffic road. The station is adjacent to Hong Chong Road, which leads to the Cross Harbour Tunnel. The traffic volume of the road is extremely high which is more than 170,000 vehicles per day.

Kwun Tong (KT): It is close to the residential buildings and most of vehicles are light- and heavy-duty vehicles. Kwun Tong belongs to one of the EPD air quality monitoring stations, which were chosen for data comparison; it represents as mixed residential/commercial/industrial area. The samples were collected on the rooftop of 25 m.

2.2. Sampling method

The monitoring program for PM$_{2.5}$ had been performed at two urban monitoring stations in Hong Kong during the two studies periods ((1) November 2000 to February 2001 and (2) June 2001 to August 2001). PM$_{2.5}$ samples were collected once every 6 days (24-h sampling duration) at PolyU and KT stations.

The high volume (hi-vol) samplers manufactured by Andersen Instruments/GMW were used for PM$_{2.5}$ sampling at two monitoring stations. The hi-vol samplers were operated at flow rates of 1.13–1.41 m^3 min^{-1}. PM$_{2.5}$ samples were collected on 20.3 cm × 25.4 cm Whatman quartz microfiber filters. The filters were pre-heated before sampling at 900 °C for 3 h. A balance for hi-vol filters (Sartorius, analytic) with accuracy of 0.1 mg was used to weigh the filter paper which was conditioned in an electronic desiccator before and after sample collection for 24 h. After collection, loaded filters were stored in a refrigerator at about 4 °C before chemical analysis to limit the evaporation of volatile components. Field blank filters were also collected to indicate the artifacts collected onto the filter before/during/after sampling.

2.3. Chemical analysis

After sampling, the filters were conditioned and weighted again to determine the mass concentration of the loaded particles. The filters were then cut into four portions for individual analysis. The filters are then analyzed with different analytical methods: (1) atomic absorption spectrophotometer (AAS) for
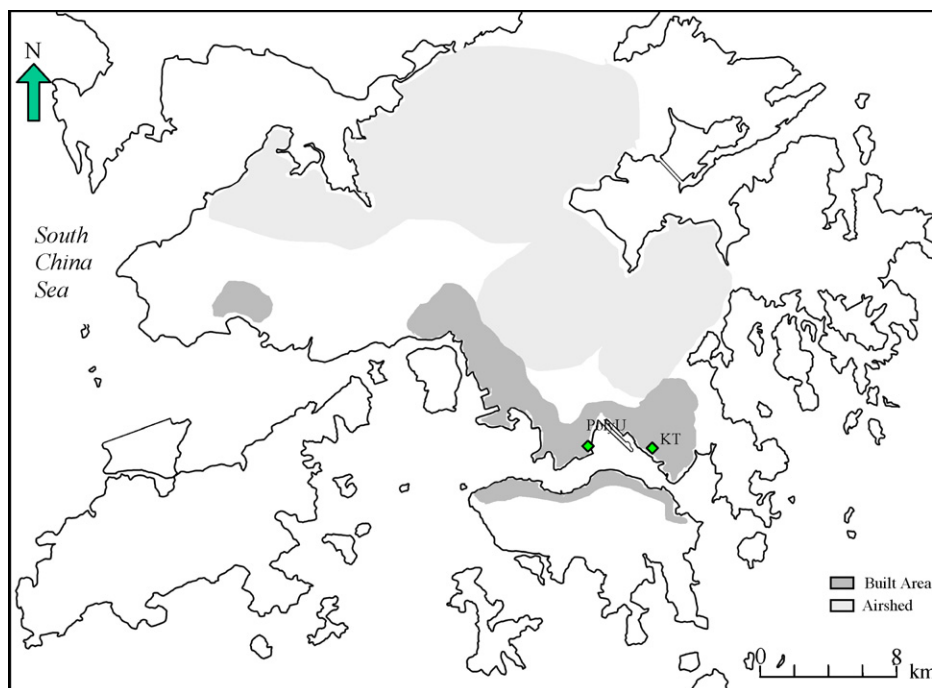

Fig. 1. Location of monitoring sites: Hong Kong Polytechnic University Campus [PolyU]; Kwun Tong (KT); Hok Tsui (HT).

Table 1
Method detection limit (MDL) of selected species determined

Analytical methods	Species	MDL
TOR	OC	$0.0958\,\mu g\,m^{-3}$
TOR	EC	$0.0958\,\mu g\,m^{-3}$
IC	Cl^-	$8.0\,ng\,m^{-3}$
IC	NO_3^-	$5.0\,ng\,m^{-3}$
IC	SO_4^{2-}	$8.0\,ng\,m^{-3}$
IC	NH_4^+	$8.0\,ng\,m^{-3}$
AAS	Na^+	$15\,ng\,m^{-3}$
AAS	K^+	$15\,ng\,m^{-3}$
ICP–MS	Al	$20\,ng\,m^{-3}$
ICP–MS	As	$1.0\,ng\,m^{-3}$
ICP–MS	Ca	$5.0\,ng\,m^{-3}$
ICP–MS	Cr	$1.0\,ng\,m^{-3}$
ICP–MS	Cu	$5.0\,ng\,m^{-3}$
ICP–MS	Fe	$5.0\,ng\,m^{-3}$
ICP–MS	Mg	$3.0\,ng\,m^{-3}$
ICP–MS	Mn	$1.0\,ng\,m^{-3}$
ICP–MS	Ni	$2.0\,ng\,m^{-3}$
ICP–MS	Pb	$1.0\,ng\,m^{-3}$
ICP–MS	Sr	$1.0\,ng\,m^{-3}$
ICP–MS	Ti	$1.0\,ng\,m^{-3}$
ICP–MS	V	$2.0\,ng\,m^{-3}$
ICP–MS	Zn	$5.0\,ng\,m^{-3}$
ICP–MS	Ba	$3.0\,ng\,m^{-3}$
ICP–MS	Cd	$0.1\,ng\,m^{-3}$
ICP–MS	Ga	$1.0\,ng\,m^{-3}$

sodium and potassium, (2) inductively coupled plasma–mass spectrometry (ICP–MS) for elements. The sample solutions were measured in triplicates, and quality controls and blanks were inserted at every 10 samples. The relative standard deviations of the measured element concentrations were typically <5%. Precision and bias were <10%. Element concentrations of the procedural blanks were generally <5% of the samples. (3) Ion chromatography (IC) for water-soluble inorganic ions. Uncertainties were ±6% for Cl^- and were ±12% for NO_3^-, SO_4^{2-} and NH_4^+. (4) Thermal/optical reflectance (TOR) method for organic carbon (OC) and elemental carbon (EC). The difference determined from replicate analyses was smaller than 5% for TC (total carbon), and 10% for OC and EC. Method detection limit (MDL) of each species was shown in Table 1. The chemical analyses were carried out by the Air Laboratory of PolyU (OC/EC and elements), Department of Chemical Engineering of Hong Kong University of Science and Technology (water-soluble ions), and Department of Chemistry of The Chinese University of Hong Kong (heavy metals).

2.4. Receptor models

Enrichment factor analysis and multivariate techniques such as correlations, absolute principal component analyses (APCA), and cluster analysis (CA) were used to define a relationship between the sources and the receptor. These analytical methods were combined to assist the identification of sources and the apportionment of the observed pollutant concentrations to those sources in the urban area of Hong Kong.

3. Results and discussion

To maximize the source-identification power of factor analysis, only 25 species were selected (after eliminating species with missing data or below detection limit). In general, one should exclude those elements from the analyses with many missing values due to poor detectability, but this must be balanced by including as many elements as possible to increase the degrees of freedom for tuning the model [12]. The 25 species are OC, EC, Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Al, As, Ca, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Sr, Ti, V, Zn, Ba, Cd and Ga. Median values were substituted for missing data. Values below detection limit (limit of detection) will be replaced by half of the minimum value reported. The average concentrations and standard deviations of the selected species were shown in Table 2. Generally speaking, the concentrations of metal levels determined in this study (both sites) were lower than other major cities in China (Table 3).

3.1. Correlations of selected species in $PM_{2.5}$

Correlations of selected species in $PM_{2.5}$ at PolyU and KT stations were determined by regression analysis. Their correlation coefficients (r) are shown in Tables 4 and 5, respectively. Nitrate (NO_3^-), sulfate (SO_4^{2-}) and ammonium (NH_4^+) are secondary pollutants, and arise from the oxidation of anthropogenic gases (NO_X, SO_2 and NH_3, respectively). Correlation coefficients among NO_3^-, SO_4^{2-} and NH_4^+ in both stations were from 0.71 to 0.91 ([**]$P < 0.01$). Ammonium nitrate and ammonium sulfate are the possible compounds in the fine aerosols due to the secondary formation from anthropogenic origin. Good correlations ($r = 0.52$–0.97, [**]$P < 0.01$) were also observed among the marking species for the crustal matter (Al, Ca, Fe, Mg and Ti) at both stations (especially in KT) even in fine particles. It implies that they should come from the same sources. At PolyU, the major sources of crustal elements came from paved road dust and they mainly appeared in the large particles (TSP or PM_{10}). Therefore, the correlation coefficients among the crustal elements were lower relative to the KT site, where the major sources of crustal elements were due to resuspension and transportation of fine crustal matter. Some heavy metals, such as Cr and Cu were also observed to have good correlations with crustal elements in KT.

At PolyU, the major emission sources of carbonaceous species came from vehicular exhaust and fuel evaporation. A fairly good correlation was observed between OC and EC ($r = 0.42$) because gasoline vehicle exhaust and fuel evaporation were the major sources of OC, however, diesel vehicle exhaust was the major source of EC. At KT, the correlation of OC and EC was slightly higher with $r = 0.65$ ([**]$P < 0.01$). From previous studies [4,7,16,17], heavy metal could be used as marker species. The correlation coefficients between Ni and V are 0.52 ([**]$P < 0.01$) in PolyU and 0.68 ([**]$P < 0.01$) in KT. It is a clear indication for combustion of oil in previous studies [16,18]. Also Pb and Cd are well correlated ($r = 0.61$, [**]$P < 0.01$) at PolyU site as they mainly came from the traffic sources. However, poor correlation ($r = 0.24$) was observed at KT because Cd might

Table 2
Average concentrations and standard deviations of selected species at PolyU and KT stations

PolyU (ng/m^3)	Mean ($n = 30$)	S.D.	KT (ng/m^3)	Mean ($n = 29$)	S.D.
	41.73[a]	12.63[a]		43.93[a]	21.08[a]
OC	8655.14	2519.25	OC	7946.02	3353.40
EC	5971.61	946.34	EC	4734.13	870.78
Cl$^-$	195.90	276.89	Cl$^-$	310.87	510.50
NO$_3^-$	1198.78	1185.93	NO$_3^-$	1459.72	1369.51
SO$_4^{2-}$	8096.91	5914.97	SO$_4^{2-}$	10042.43	6205.26
Na$^+$	493.72	260.62	Na$^+$	937.66	987.72
NH$_4^+$	1884.63	1443.19	NH$_4^+$	1779.44	1090.25
K$^+$	564.40	669.69	K$^+$	649.02	580.24
Al	119.40	92.98	Al	226.84	281.50
As	2.65	2.73	As	3.01	3.49
Ca	349.36	227.10	Ca	531.25	572.11
Cr	1.57	1.39	Cr	2.76	3.31
Cu	17.04	6.91	Cu	28.64	17.56
Fe	191.41	101.95	Fe	325.33	393.12
Mg	91.14	58.01	Mg	156.54	181.36
Mn	10.91	6.73	Mn	22.55	16.02
Ni	8.05	4.46	Ni	7.98	5.57
Pb	42.79	40.18	Pb	56.72	51.88
Sr	3.39	2.79	Sr	2.28	1.29
Ti	1.93	2.59	Ti	6.37	10.51
V	7.29	5.38	V	6.12	5.58
Zn	166.95	179.04	Zn	230.23	313.33
Ba	22.33	24.70	Ba	35.29	45.24
Cd	0.97	0.46	Cd	1.47	0.77
Ga	2.60	4.19	Ga	4.80	6.73

[a] Mass (μg/m^3).

come from other industrial activities also [19]. As is a tracer for coal combustion and Zn is a good marker for tire wear and non-ferrous smelters. It implies that the combination of carbonaceous compounds (OC and EC) with inorganic species would give more detail information.

3.2. Enrichment factor of elements PM$_{2.5}$

Enrichment factors (EF) of trace elements in PM$_{2.5}$ relative to the earth's crust were calculated to indicate the extent of contribution of sources other than natural crust to the ambient elemental levels [20,22]. In this study, Fe was used as reference, and the compositions of the earth's crust were taken from Mason and Moore (1982) [21]. Trace elements EFs include some degree of uncertainty related to the natural variations of the earth crustal composition. For this reason, it is usually assumed that the EFs should be more than an order of magnitude higher than unity to suggest an anthropic origin [22,23].

Results given in Table 6 showed that Al, Ti, Mg, Ca and K have small enrichment factors since they are mostly crustal. However, Cd is the most enriched elements in PM$_{2.5}$, (average = 1794 and 1624, at PolyU and KT, respectively) followed by Pb, Zn, As and Cu. For these elements non-crustal sources such as vehicular exhaust and industrial emission may be suggested. Nevertheless, enrichment of crustal components in fine particles can also occur as a result of their transport to some distance before being removed from the atmosphere by deposition processes [10]. Mn, Cr and Sr showed low EF which

suggested that these crustal sources also predominate in PM$_{2.5}$. For these elements, natural emissions are very important and normally exceed anthropogenic sources. Lower EF values with increasing particle size have been reported for Cd, Pb, Zn and Ni [24–26]. On the other hand, Eleftheriadis and Colbeck (2001), found increasing EFs with size for coarse V, Cu and Cr with highest enrichment at around 10 μm and at the very large sizes [27].

3.3. Principal component analysis (PCA)/absolute principal component analysis (APCA) of selected species in PM$_{2.5}$

PCA has been used generally as an exploratory tool to identify the major sources of aerosol emissions and to statistically select independent source tracers [16]. Result of varimax rotated factor analysis carried out on various selected ambient air components at PolyU and KT and the corresponding possible sources are depicted in Tables 7 and 8, respectively. Six factors at each site were isolated based on the following criteria. Firstly, the number of factors were selected such that the cumulative percentage variance explained by all the chosen factors is more than 80%. Secondly, only the factors with eigenvalue more than one were chosen. Since higher factor loading of particular elements (marker elements) in a factor can help identify the possible sources [28], the number of factors selected (sources identified) should represent the sources which are relevant in the receptor domain [29].

Table 3
Comparison of concentrations of metal levels at the major cities of China

	PolyU	KT	Beijing [33]	Beijing [34]	Shanghai [35]	Guangzhou [36]		Wuhan [36]		Lanzhou [36]		Chongqing [36]	
	$PM_{2.5}$[a]	$PM_{2.5}$[a]	PM_{10}[a]	TSP[a]	PM_{10}[a]	$PM_{2.5}$[a]	$PM_{2.5-10}$[a]	$PM_{2.5}$[a]	$PM_{2.5-10}$[a]	$PM_{2.5}$[a]	$PM_{2.5-10}$[a]	$PM_{2.5}$[a]	$PM_{2.5-10}$[a]
			2001–2003	1998	2001–2002	1995–1996		1995–1996		1995–1996		1995–1996	
Al	119.4	226.84	3500										
As	2.65	3.01	48		42.1	36.8	7.3	25.5	3.75	34.25	8.3	30.55	4.7
Ca	349.36	531.25			9634								
Cr	1.57	2.76	19		32.3								
Cu	17.04	28.64	110	117.74	171	47.3	28.85	44.95	24.55	28.1	18	18.45	8.2
Fe	191.41	325.33	5500		2690								
Mg	91.14	156.54			1984.00								
Mn	10.91	22.55	240		186.00								
Ni	8.05	7.98	22		13.9								
Pb	42.79	56.72	430	235.24	515	473	86.65	257.5	26.9	524	103.2	212	61.2
Sr	3.39	2.28			33.2								
Ti	1.93	6.37	470		221								
V	7.29	6.12	13		17.6								
Zn	166.95	230.23	770	365.90	1409	622	206	264.5	66.7	640.5	118.55	228.5	85.6
Ba	22.33	35.29			2479								
Cd	0.97	1.47	6.8	2.61	10.9								
Ga	2.6	4.8											

Unit: ng/m^3.

[a] Size.

Table 4
Summary of correlation coefficients of selected species in PM$_{2.5}$ at PolyU stations

PolyU	OC	EC	Cl⁻	NO₃⁻	SO₄²⁻	Na⁺	NH₄⁺	K⁺	Al	As	Ca	Cr	Cu	Fe	Mg	Mn	Ni	Pb	Sr	Ti	V	Zn	Ba	Cd	Ga
OC	1.00	0.42	0.00	0.38	0.41	0.23	0.39	0.50	0.28	0.52	0.03	0.21	0.63	0.25	0.19	0.25	−0.06	0.57	0.19	0.20	−0.05	0.33	0.13	0.28	0.25
EC		1.00	0.05	0.04	0.07	−0.31	−0.23	−0.11	−0.21	0.08	−0.21	−0.39	0.27	−0.27	−0.19	0.05	−0.04	−0.03	−0.01	−0.31	0.12	0.06	0.07	0.01	0.13
Cl⁻			1.00	0.43	0.52	−0.03	0.05	0.00	0.12	−0.05	0.00	−0.05	−0.16	0.00	0.25	0.03	−0.05	0.09	0.20	0.03	−0.19	0.23	0.10	−0.20	0.08
NO₃⁻				1.00	0.83	0.40	0.76	0.73	0.30	0.56	0.15	0.40	0.07	0.36	0.22	−0.11	−0.47	0.81	0.55	0.45	−0.32	0.64	0.15	0.28	0.18
SO₄²⁻					1.00	0.43	0.72	0.69	0.48	0.53	0.27	0.30	0.07	0.44	0.38	0.01	−0.36	0.70	0.59	0.46	−0.25	0.66	0.35	0.32	0.32
Na⁺						1.00	0.68	0.52	0.64	0.26	0.43	0.44	0.08	0.69	0.44	0.35	−0.20	0.48	0.59	0.57	−0.38	0.55	0.23	0.07	0.40
NH₄⁺							1.00	0.78	0.52	0.60	0.47	0.63	0.06	0.63	0.37	0.00	−0.39	0.80	0.49	0.68	−0.30	0.53	0.13	0.40	0.14
K⁺								1.00	0.59	0.80	0.19	0.62	0.28	0.60	0.46	0.05	−0.35	0.94	0.40	0.67	−0.36	0.55	0.25	0.58	0.25
Al									1.00	0.45	0.54	0.45	0.13	0.65	0.85	0.45	0.21	0.49	0.42	0.70	−0.30	0.50	0.11	0.26	0.46
As										1.00	0.21	0.32	0.38	0.30	0.38	−0.03	−0.14	0.77	0.24	0.49	−0.13	0.42	0.08	0.66	0.21
Ca											1.00	0.31	0.12	0.52	0.52	0.23	0.11	0.14	0.22	0.54	−0.14	0.22	−0.03	0.03	−0.01
Cr												1.00	0.05	0.63	0.40	0.01	−0.20	0.61	0.13	0.71	−0.20	0.12	−0.16	0.33	−0.12
Cu													1.00	0.29	0.19	0.17	0.11	0.26	0.05	0.21	0.19	0.16	0.06	0.16	0.04
Fe														1.00	0.65	0.31	−0.21	0.50	0.26	0.87	−0.25	0.39	0.15	0.18	0.08
Mg															1.00	0.54	0.35	0.31	0.09	0.75	−0.14	0.21	−0.07	0.10	0.02
Mn																1.00	0.29	−0.05	−0.04	0.25	−0.05	0.04	−0.02	−0.45	0.09
Ni																	1.00	−0.37	−0.17	−0.10	0.52	−0.23	−0.20	−0.06	−0.13
Pb																		1.00	0.48	0.55	−0.33	0.62	0.27	0.61	0.31
Sr																			1.00	0.25	−0.22	0.92	0.32	0.29	0.68
Ti																				1.00	−0.19	0.35	−0.18	0.30	−0.04
V																					1.00	−0.31	−0.27	0.08	−0.32
Zn																						1.00	0.38	0.38	0.73
Ba																							1.00	0.15	0.43
Cd																								1.00	0.21
Ga																									1.00

(Data highlight: $P < 0.01$).

Table 5
Summary of correlation coefficients of selected species in PM$_{2.5}$ at KT stations

KT	OC	EC	Cl⁻	NO₃⁻	SO₄²⁻	Na⁺	NH₄⁺	K⁺	Al	As	Ca	Cr	Cu	Fe	Mg	Mn	Ni	Pb	Sr	Ti	V	Zn	Ba	Cd	Ga
OC	1.00	0.65	0.25	0.64	0.76	0.25	0.85	0.84	0.41	0.83	0.26	0.46	0.40	0.36	0.25	0.11	−0.03	0.81	0.25	0.32	0.03	0.51	0.38	0.23	0.44
EC		1.00	0.09	0.34	0.43	0.14	0.44	0.40	0.14	0.31	0.20	0.17	0.30	0.23	0.14	0.12	−0.21	0.50	0.15	0.15	0.02	0.40	0.53	0.00	0.49
Cl⁻			1.00	0.76	0.47	0.94	0.27	0.41	0.89	0.24	0.80	0.69	0.80	0.78	0.88	0.31	0.18	0.24	0.22	0.87	−0.14	0.15	0.22	−0.12	0.08
NO³⁻				1.00	0.85	0.67	0.71	0.78	0.70	0.57	0.54	0.66	0.63	0.59	0.58	0.02	−0.09	0.56	0.19	0.62	−0.22	0.32	0.46	−0.17	0.26
SO₄²⁻					1.00	0.37	0.91	0.85	0.45	0.64	0.32	0.59	0.45	0.37	0.29	−0.20	−0.22	0.65	0.24	0.35	−0.23	0.38	0.61	−0.11	0.37
Na⁺						1.00	0.22	0.39	0.89	0.23	0.83	0.66	0.79	0.80	0.93	0.38	0.24	0.22	0.24	0.88	−0.05	0.11	0.07	−0.10	0.02
NH₄⁺							1.00	0.94	0.34	0.78	0.17	0.50	0.31	0.25	0.15	−0.24	−0.22	0.71	0.16	0.22	−0.27	0.39	0.46	−0.03	0.33
K⁺								1.00	0.51	0.79	0.28	0.56	0.43	0.39	0.29	−0.04	−0.21	0.76	0.13	0.37	−0.36	0.46	0.36	0.00	0.40
Al									1.00	0.40	0.83	0.64	0.83	0.83	0.91	0.49	0.32	0.39	0.18	0.90	−0.06	0.28	0.19	0.08	0.26
As										1.00	0.28	0.49	0.33	0.40	0.27	0.16	0.05	0.79	0.35	0.37	−0.08	0.40	0.12	0.25	0.26
Ca											1.00	0.79	0.94	0.97	0.95	0.54	0.30	0.38	0.58	0.96	0.01	0.26	0.15	−0.04	0.23
Cr												1.00	0.80	0.85	0.71	0.27	0.11	0.58	0.69	0.79	−0.12	0.35	0.16	−0.14	0.22
Cu													1.00	0.93	0.87	0.48	0.25	0.50	0.53	0.90	−0.05	0.40	0.30	0.02	0.39
Fe														1.00	0.91	0.61	0.24	0.53	0.63	0.97	−0.05	0.41	0.13	−0.03	0.33
Mg															1.00	0.51	0.39	0.25	0.37	0.96	0.05	0.12	0.06	0.00	0.07
Mn																1.00	0.37	0.32	0.32	0.61	0.16	0.54	−0.13	0.26	0.44
Ni																	1.00	−0.09	0.26	0.29	0.68	−0.16	−0.21	0.49	−0.20
Pb																		1.00	0.51	0.41	−0.23	0.72	0.41	0.24	0.67
Sr																			1.00	0.48	0.21	0.29	0.09	0.03	0.20
Ti																				1.00	−0.03	0.35	0.08	−0.04	0.22
V																					1.00	−0.27	−0.20	0.34	−0.28
Zn																						1.00	0.38	0.08	0.82
Ba																							1.00	0.00	0.63
Cd																								1.00	0.13
Ga																									1.00

(Data highlight: $P < 0.01$).

Table 6
Enrichment factors of major elements and heavy metals at three sampling sites

Species	PolyU	KT
Al	0.268	0.293
Ca	2.129	1.905
Ti	0.118	0.228
Mg	1.253	1.266
K	1.609	1.102
Na	6.580	4.579
Zn	429.597	332.283
As	338.189	205.280
Cr	0.888	0.813
Cu	124.656	123.236
Mn	3.324	4.044
Ni	73.599	44.624
Pb	391.229	305.096
Sr	1.710	0.702
V	22.223	10.978
Ba	13.237	12.938
Cd	1793.881	1324.261
Ga	27.439	29.185

At PolyU, six factors were determined, summing 80% of the total variance in the fine particle data set. Factor 1 explains 21.9% of the variance and presented high loading for Al, Ca, Fe, Mg and Ti; thus it can be interpreted as crustal contribution. This can be proved by the low enrichment factors (EF) of these elements. A significant amount of road dust is present near PolyU and is also kept in suspension by vehicular movement. OC, Pb, Zn and Cr were observed in paved road dust profiles of PolyU

[30] which have moderate to minor loading in factor 1. Hence, this factor can be identified as the road dust component. Factor 2 explains 18.5% of the variance and contains high loading of K^+, As, Pb and Cd. These elements are used as markers for non-ferrous metal smelter and automobile emission [4,5,12]. Moderate loading of OC, NO_3^-, SO_4^{2-} and NH_4^+ were also observed in factor 2. They are secondary aerosols and the by-products of combustion. Hence, this factor can be identified as automobile emission plus secondary aerosol. Factor 3 is heavily loaded with Zn, Sr and Ga with percentage variance of 13.5. Harrison et al. (1996) and Manoli et al. (2002) used Zn as the marker for tire wear [7,16]. Incineration is also the possible source for Zn [31]. However, there are no significant sources for incineration around PolyU. Therefore, this factor can be determined as tire wear. Factor 4 explains 9.3% of the variance and was highly loaded with V and Ni. The correlation coefficient between Ni and V is high (see previous section). As discussed before, V and Ni are marker species for oil combustion [16,18]. Thus, factor 4 is identified as oil combustion source. Factor 5 has a high factor loading for OC, EC and Cu with percentage variance of 9.1. It is obvious that OC and EC come from vehicular exhaust (especially at PolyU site). Also diesel engines are the source of Cu [7]. This factor was interpreted as representing emissions from diesel vehicles. Finally, high to moderate loading for Cl^-, NO_3^-, SO_4^{2-} with percentage variance of 7.6 which indicated the presence of secondary or transportation aerosols in factor 6.

Six factors were also obtained at KT station with eigenvalues >1, explaining 90.9% of the total variance. Basically the six

Table 7
The results of principal component analysis for selected species in PM$_{2.5}$ at PolyU stations (varimax with Kaiser normalization)

Pollutants	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Communality
OC	0.129	0.311	0.140	0.099	0.823	0.057	0.824
EC	−0.412	−0.079	0.092	−0.077	0.666	0.184	0.668
Cl^-	0.025	−0.095	0.076	0.023	−0.070	0.924	0.875
NO_3^-	0.174	0.587	0.199	0.397	0.114	0.561	0.900
SO_4^{2-}	0.264	0.526	0.348	0.221	0.134	0.599	0.893
Na^+	0.685	0.103	0.450	0.346	−0.021	−0.079	0.809
NH_4^+	0.509	0.624	0.159	0.386	0.003	0.160	0.849
K^+	0.438	0.714	0.191	0.320	0.249	0.068	0.907
Al	0.833	0.174	0.368	−0.132	0.082	0.099	0.893
As	0.244	0.743	0.114	−0.014	0.377	0.061	0.771
Ca	0.684	0.051	0.055	−0.065	−0.089	0.024	0.486
Cr	0.570	0.514	−0.232	0.300	−0.111	−0.081	0.751
Cu	0.150	0.172	−0.004	−0.124	0.755	−0.205	0.680
Fe	0.802	0.231	0.067	0.301	0.097	−0.056	0.803
Mg	0.871	0.066	−0.033	−0.224	0.141	0.258	0.901
Mn	0.588	−0.521	0.046	−0.036	0.418	0.060	0.799
Ni	0.200	−0.247	−0.082	−0.871	0.041	−0.052	0.871
Pb	0.296	0.760	0.255	0.327	0.258	0.150	0.926
Sr	0.179	0.293	0.807	0.054	−0.080	0.208	0.821
Ti	0.831	0.416	−0.102	0.116	0.013	0.064	0.892
V	−0.207	0.061	−0.286	−0.689	0.087	−0.108	0.622
Zn	0.228	0.358	0.797	0.102	0.090	0.237	0.891
Ba	−0.100	0.009	0.610	0.242	0.110	−0.011	0.453
Cd	−0.021	0.868	0.203	−0.215	0.022	−0.185	0.875
Ga	0.037	0.027	0.909	0.020	0.099	−0.019	0.839
% of variance	21.916	18.539	13.451	9.340	9.107	7.648	
Cumulative %	21.916	40.455	53.906	63.246	72.352	80.000	

Table 8
The results of principal component analysis for selected species in $PM_{2.5}$ at KT stations (varimax with Kaiser normalization)

Pollutants	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Communality
OC	0.140	0.860	0.197	0.184	0.298	0.069	0.925
EC	0.047	0.310	0.216	0.054	0.711	0.103	0.664
Cl^-	0.943	0.168	−0.072	−0.090	0.083	−0.081	0.944
NO_3^-	0.611	0.631	−0.093	−0.191	0.279	−0.047	0.896
SO_4^{2-}	0.283	0.790	−0.097	−0.204	0.416	0.072	0.932
Na^+	0.952	0.137	−0.066	−0.002	0.009	−0.052	0.932
NH_4^+	0.100	0.931	−0.027	−0.161	0.244	0.036	0.963
K^+	0.263	0.906	0.118	−0.188	0.121	−0.070	0.959
Al	0.912	0.273	0.152	0.108	0.037	−0.162	0.969
As	0.151	0.891	0.205	0.145	−0.121	0.148	0.917
Ca	0.909	0.058	0.153	0.056	0.075	0.316	0.963
Cr	0.681	0.412	0.061	−0.131	−0.003	0.515	0.919
Cu	0.855	0.182	0.220	0.043	0.215	0.242	0.919
Fe	0.864	0.167	0.289	0.015	0.026	0.358	0.986
Mg	0.970	0.073	0.028	0.136	−0.003	0.090	0.973
Mn	0.462	−0.190	0.738	0.302	−0.131	0.101	0.912
Ni	0.309	−0.134	−0.071	0.824	−0.160	0.090	0.832
Pb	0.164	0.722	0.534	−0.002	0.186	0.283	0.948
Sr	0.280	0.140	0.160	0.137	0.030	0.896	0.947
Ti	0.938	0.145	0.214	0.038	−0.033	0.192	0.986
V	−0.043	−0.181	−0.253	0.832	0.084	0.236	0.853
Zn	0.119	0.319	0.808	−0.144	0.234	0.116	0.858
Ba	0.080	0.230	0.118	−0.140	0.867	−0.059	0.847
Cd	−0.107	0.166	0.314	0.752	−0.073	−0.209	0.751
Ga	0.064	0.204	0.775	−0.125	0.509	0.030	0.922
% of variance	32.980	22.283	10.753	9.448	8.604	6.807	
Cumulative %	32.980	55.264	66.017	75.464	84.068	90.875	

Extraction method: principal component analysis. Rotation method: varimax with Kaiser normalization.

factors are very similar to that obtained at PolyU; but in each factor it may contain more than one source. Factor 1 explains 33.0% of the variance which represented crustal matter and some secondary aerosols (NO_3^- and SO_4^{2-}). Hence, this factor can be identified as the crustal matter component plus secondary aerosols. Factor 2 explains 22.3% of the variance and contains high loading of K^+, As, Pb and Cd, which are markers for automobile emission and non-ferrous metal smelter. Moreover, high loading of OC, NO_3^-, SO_4^{2-} and NH_4^+ was also observed in factor 2. Other than combustion, this source type could be the result of the continuous oxidation of primary pollutants (VOCs, SO_2, NO_X and NH_3) taking place during atmospheric transport. Hence, this factor can be identified as automobile emission plus secondary aerosols. Factors 3 and 4 explain 10.8 and 9.4% of the variance, respectively. As in PolyU, factor 3 is identified as tire wear, while factor 4 for oil combustion source. Factor 5 has a high factor loading for EC and Ba with percentage variance of 9.1%. This factor was identified as emissions from diesel vehicles. Finally, the last factor explains 6.8% of the variance and contains high to moderate loading of crustal elements (Sr, Ca and Fe) and heavy metals (Cr, Cu, Pb and V). This factor was interpreted as emissions from several metallurgical activities along with crustal matter occurring in the industrial area.

APCA can provide a quantitative elemental source profile, instead of just a qualitative factor-loading matrix as in PCA. Using the grouping results of PCA in the previous section, source contributions were then calculated using multiple regression of particle mass concentration on absolute principal component scores (APCS). Regressing the gravimetric data on these APCS can give estimates of the coefficients that convert the APCS into mass contributions from each source for each sample. For each source identified by the APCA, the weighted regression of each element's concentration on the predicted mass contributions yields estimates of the content of that element in each source. Tables 9 and 10 present the APCA source apportionment of $PM_{2.5}$ in PolyU and KT given in the original concentration units (ng/m^3), respectively. The estimated quantitative source apportionments for $PM_{2.5}$ are shown in Figs. 2 and 3, respectively. Diesel emission is responsible for the most of the $PM_{2.5}$ mass at PolyU station (about 47%); and automobile emission plus non-ferrous metal smelter also composed about 15% of the $PM_{2.5}$ fraction. Secondary aerosols are the second most important factor (about 18%) at PolyU. Crustal matter also dominates the $PM_{2.5}$ fraction (about 6% at PolyU). However, at KT, some factor represents more than one source. For example, automobile emission plus non-ferrous metal smelter mixed with secondary aerosols, which composed about 44% of the total $PM_{2.5}$ mass. Secondary aerosols formed during transportation were due to the primary emission of SO_2 and NO_X from vehicular emission. About 30% of $PM_{2.5}$ was also calculated as crustal matter and secondary aerosols. Diesel emission contributed 14% of the total $PM_{2.5}$ mass. The large unexplained fractions in PolyU and KT maybe ascribed to the water uptake in the quartz filters [32]

Table 9
The Results of APCA estimated concentration for selected species in $PM_{2.5}$ at PolyU stations

Factor concentrations (ng/m^3)

Pollutants	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Sum of estimated contributions
APCA estimated							
OC	0.0	0.0	0.0	0.0	10749.7	0.0	9485.8
EC	0.0	0.0	4.1	0.0	3557.6	387.6	2555.6
Cl^-	0.0	0.0	0.0	3.7	0.0	560.6	135.4
NO_3^-	0.0	926.1	0.0	0.0	151.0	1143.3	1234.5
SO_4^{2-}	0.0	3804.3	343.6	22.4	80.5	6175.2	9283.3
Na^+	326.5	0.0	223.0	0.0	0.0	0.0	101.5
NH_4^+	615.2	1238.0	0.0	0.0	0.0	91.1	783.6
K^+	124.3	705.9	0.0	0.0	482.2	0.0	1076.3
Al	145.6	0.0	65.2	3.0	0.0	0.4	162.6
As	0.0	4.4	0.0	0.1	3.3	0.0	6.5
Ca	331.0	0.0	14.1	3.6	0.0	0.0	96.1
Cr	1.2	1.3	0.0	0.0	0.0	0.0	−0.5
Cu	0.9	0.0	0.0	0.0	7.5	0.0	23.5
Fe	150.6	0.0	0.0	0.0	36.2	0.0	86.7
Mg	104.3	0.0	0.0	2.1	16.6	31.1	112.2
Mn	10.9	0.0	0.3	0.0	18.1	0.6	14.4
Ni	3.8	0.7	1.6	0.4	0.0	0.8	5.9
Pb	0.0	49.4	0.0	0.0	28.0	0.7	66.2
Sr	0.0	0.7	4.9	0.1	0.0	0.3	2.0
Ti	3.7	1.2	0.0	0.0	0.0	0.0	2.2
V	0.0	6.6	0.0	0.4	0.4	0.5	4.8
Zn	0.0	36.4	286.2	2.8	0.0	23.4	259.6
Ba	0.0	0.0	34.7	0.0	10.7	0.0	12.3
Cd	0.0	1.3	0.2	0.0	0.0	0.0	0.4
Ga	0.0	0.0	9.4	0.1	0.0	0.0	4.7

Table 10
The results of APCA estimated concentration for selected species in $PM_{2.5}$ at KT stations

Factor concentrations (ng/m^3)

| Pollutants	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Sum of estimated contributions
APCA estimated							
OC	0.0	7185.8	0.0	1853.1	965.0	0.0	7788.0
EC	0.0	0.0	0.0	316.8	2696.9	177.3	2239.7
Cl^-	919.9	0.0	0.0	0.0	129.1	0.0	275.3
NO_3^-	1189.3	1457.4	0.0	0.0	749.0	0.0	1643.3
SO_4^{2-}	110.0	9201.3	0.0	0.0	6353.7	706.8	10287.4
Na^+	1754.1	0.0	0.0	0.0	0.0	0.0	530.5
NH_4^+	0.0	2700.9	0.0	0.0	0.0	0.0	1460.2
K^+	0.0	1449.8	21.1	0.0	0.0	0.0	544.5
Al	461.7	69.1	63.9	60.6	0.0	0.0	271.3
As	0.0	10.8	0.9	1.0	0.0	0.4	4.1
Ca	692.3	0.0	0.0	0.0	173.0	294.2	635.8
Cr	1.2	1.9	0.0	0.0	0.0	4.4	2.9
Cu	19.4	0.0	1.1	0.4	13.3	4.5	26.8
Fe	373.6	0.0	139.5	0.0	0.0	235.2	410.4
Mg	292.5	0.0	0.0	29.1	6.2	0.0	164.9
Mn	8.3	0.0	33.0	4.2	0.0	0.0	10.2
Ni	2.2	0.8	0.0	7.8	0.5	0.0	8.5
Pb	0.0	78.4	50.7	0.4	0.0	25.5	88.7
Sr	0.0	0.0	0.0	0.0	0.1	3.5	1.9
Ti	13.9	0.0	3.0	0.0	0.0	0.4	8.3
V	0.0	0.0	0.0	8.5	9.0	3.9	11.6
Zn	0.0	0.0	630.7	0.0	0.0	0.0	314.7
Ba	6.1	0.0	0.0	5.2	178.9	0.0	105.1
Cd	0.0	0.9	0.7	1.1	0.0	0.0	1.2
Ga	0.0	0.0	12.0	0.0	9.9	0.0	10.7
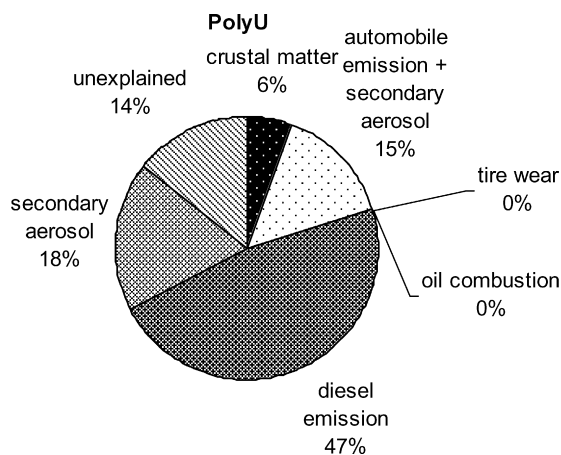
Fig. 2. Estimated source contribution to PM$_{2.5}$ at PolyU.
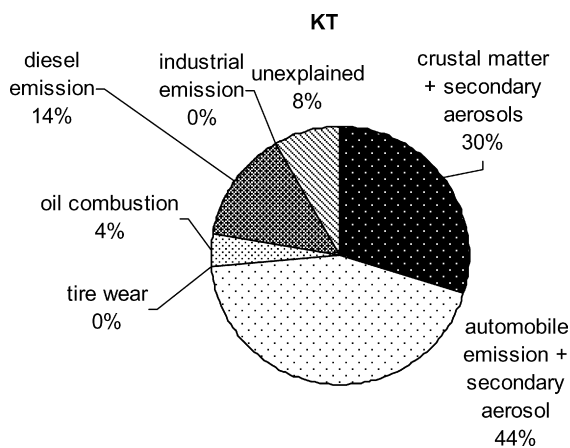

Fig. 3. Estimated source contribution to PM$_{2.5}$ at KT.

and some sources that have not been identified in the analysis. This is reasonable because of the large surface area and moisture absorption property of 20.3 cm × 25.4 cm Whatman quartz microfiber filters were used in this study [32]. This unexplained fractions were also observed in the mass closure analysis at both stations.

3.4. Cluster analysis of selected species in PM$_{2.5}$

In order to achieve a greater confidence in the final classification, cluster analysis was used for comparison purpose. Resulting dendograms (obtained through Ward's method) are observed in Figs. 4 and 5, for the PolyU and KT sites,

respectively, as obtained using SPSS 8.0 software. The results of cluster analysis in both sites agree with those of PCA. At PolyU site, there is a clear grouping of the elements K$^+$, Pb, NH$_4^+$, As, NO$_3^-$, SO$_4^{2-}$ and Cd which are commonly associated with non-ferrous metal smelter and gasoline emission. NH$_4^+$, NO$_3^-$, and SO$_4^{2-}$ are also the major secondary aerosol components in the atmosphere. The clusters are similar to the factors 2 and 6 of PCA result at PolyU. Moreover, the elements Sr, Zn, Ga, Ba and Cl$^-$ also form a group, suggesting a tire wear or incineration origin (like factor 3 in PCA). On the other hand, a cluster typically identified as crustal matter is formed by Fe, Ti, Cr, Na, Al, Mg, Ca and Mn. These elements are associated with pave road dust (like factor 1 in PCA). In addition, Ni and V form a

Fig. 4. Cluster analysis of selected species in PM$_{2.5}$ of PolyU station.

```
Dendrogram using Ward Method

                        Rescaled Distance Cluster Combine

        C A S E       0         5        10        15        20        25
       Species  Num   +---------+---------+---------+---------+---------+
       (Group)
       Ca        11   -+
       Fe        14   -+
       Ti        20   -+
       Mg        15   -+---+
       Cu        13   -+   +-------+                                    
       Cl         3   -+ I         I
       Na         6   -+---+       +--------------------------+
       Al         9   -+       I                             I
       Cr        12   ---+---------+                  +-------+
       Sr        19   ---+                    I         I
       Ni        17   -----+---+              I         I
       V         21   -----+   +------------------------+     I
       Cd        24   ---------+                              I
       OC         1   -+-+                                    I
       As        10   -+ +---+                                I
       Pb        18   ---+   I                                I
       NH4+       7   -+     +----------------+               I
       K          8   -+-+ I              I                   I
       SO4 2-     5   -+ +---+            +-----------------------+
       NO3-       4   ---+               I
       EC         2   -------+-----+     I
       Ba        23   -------+     +----------+
       Zn        22   ---+-----+ I
       Ga        25   ---+     +---+
       Mn        16   ---------+
```
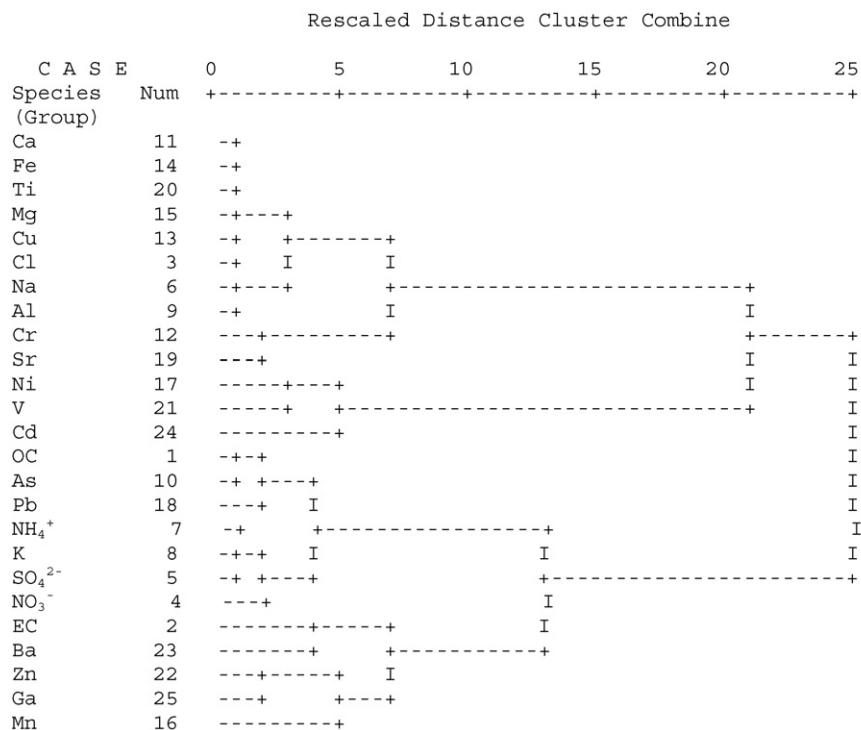
Fig. 5. Cluster analysis of selected species in PM$_{2.5}$ of KT station.

group which is suggested as oil combustion source (like factor 4 in PCA). A final group, easily identified with traffic (especially diesel engine), is based on OC, EC and Cu (like factor 5 in PCA).

The KT site dendogram presents a grouping of crustal elements, such as Ca, Fe, Ti, Mg, Cu, Cl, Na, and Al, which is similar to the PCA result for crustal matter in factor 1. To a less extent, Cr and Sr are also correlated to this group. It means the emitted heavy metal (Cr) from industrial area may be attached to the crustal matter and be transported to the receptor site (like factor 6 in PCA). On the other hand, a cluster typically identified as oil combustion is formed by Ni, V and Cd (like factor 4 in PCA). Moreover, the elements OC, As, Pb, NH$_4^+$, K$^+$, NO$_3^-$ and SO$_4^{2-}$ also form a group, which is identified as automobile and non-ferrous metal smelter emission mixed with secondary aerosols (like factor 2 in PCA). EC and Ba form a group that is suggested to be as diesel engine related species (like factor 5 in PCA). Finally, the elements Zn, Ga and Mn form a group, (undetermined), suggesting a tire wear origin (like factor 3 in PCA). It has been shown that how cluster analysis match with PCA for the identification of pollutants sources in PM$_{2.5}$.

4. Conclusion

A monitoring program for PM$_{2.5}$ had been performed at two urban monitoring stations in order to identify the main sources influencing the PM$_{2.5}$ quality in Hong Kong. Enrichment factors and correlation analysis were used as the first step to gain insight on the data and to simplify the chemical interpretation. Then PCA/(APCA and cluster analysis, were used to carry out the

source apportionment. Twenty-five chemical species in PM$_{2.5}$ were determined and selected for receptor models. Six factors at each site were isolated by using PCA/APCA analysis. Fine particulate matter produced by the transport activities is the main air pollution problem in Hong Kong. At PolyU, automobile emission plus secondary aerosol and diesel vehicles are responsible for 62% of the PM$_{2.5}$ mass. Basically the six factors in KT station are very similar to that obtained at PolyU, but in each factor it may contains not only one source. Factor 2 identified as automobile emission plus secondary aerosols which composed about 44% of the total PM$_{2.5}$ mass. Also 30% of PM$_{2.5}$ was calculated as crustal matter and secondary aerosols in factor 1. However, the large unexplained fractions in PolyU and KT maybe ascribed to the water uptake in the quartz filters and some sources that have not been identified in the analysis. Cluster analysis was used for comparison purpose in source apportionment. The results in both sites agree with the PCA results for the identification of pollutants sources in PM$_{2.5}$.

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