



Temporal Distribution and Chemical Characterization of Atmospheric Particulate Matter in the Eastern Coast of Peninsular Malaysia

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ABSTRACT

This study aims to determine the temporal distribution and composition of suspended particulate matter in different sizes PM_{2.5-10} (Coarse Particle, CP) and PM_{2.5} (Fine Particle, FP) for source apportionment analysis. CP and FP samples were collected using the Gent PM₁₀ stacked filter unit (SFU) sampler at an urban coastal city of Kuala Terengganu, in Malaysia, from August 2006 to December 2007. Samples were analyzed for elemental concentrations using Inductive Coupled Plasma-Mass Spectrometry (ICP-MS) and Inductive Coupled Plasma-Optical Emission Spectrometry (ICP-OES). Water-soluble ionic species were determined using Ion Chromatography. The results show that the concentration range of the CP and FP fractions varied from 1.17 to 28.45 µg/m³ and from 3.82 to 35.58 µg/m³ with mean concentrations (n = 80) of 10.41 ± 5.40 µg/m³ and 14.31 ± 6.53 µg/m³, respectively. On average, the measured chemical species accounted ca. 54% of the coarse particles and 32% of the fine ones. Na⁺, SO₄²⁻, Cl⁻, NO₃⁻, Al, Ca, Fe, K and Na were mainly in the coarse particle size range and accounted for ca. 52% of the coarse particle mass, while SO₄²⁻, NH₄⁺, K⁺, Al, Ca, K and Na were mainly in fine particle size range and accounted for ca. 28% of the fine particle mass. The data sets were then analyzed using Principle Component Analysis (PCA) to identify the possible sources of these fine and coarse particles based on their chemical species. For both fractions, the sources were soil dust, marine aerosol, vehicle exhaust, secondary aerosol, road dust and biomass burning. In addition, crustal enrichment factors (cEF) were calculated, and the results were consistent with those of the PCA analysis.

Keywords: Southern South China Sea; Fine and coarse particles; Principal component analysis; Enrichment factor; Source apportionment.

INTRODUCTION

Air pollution is of great concern in many countries globally due to its potential impact on health and environmental quality. Amongst air pollutants, air particulate matter (APM) is of particular concern with fine particles receiving worldwide attention due to their ability to cause adverse health effects such as asthma and potentially mortality (Dockery and Pope, 1994; Afroz *et al.*, 2003; Kappos *et al.*, 2004). These particles can also scatter and absorb incoming solar radiation thereby reducing visibility (Chun

and Lim, 2004). This may result in affecting the Earth's climate directly (by scattering and absorbing radiation) and indirectly (by serving as nuclei for cloud formation) (Svensmark *et al.*, 2009). In addition, the fine particles provide sites for surface chemistry and interactions between gases and particles (Ding and Zhu, 2003).

The majority of Asian countries have undergone rapid economic development during the last decade. Increased urbanisation, industrialization and vehicular usage in these cities, coupled with trans-boundary haze pollution and Asian dust (AD) phenomenon, has contributed to the increase in air particulate matter concentrations in Asian cities (Kim Oanh *et al.*, 2006). It is therefore not surprising that research on particulate air pollution, on spatial distribution and characterization and of source, has gain momentum in the last decade across Asia (Ho *et al.*, 2006; Ebihara *et al.*, 2008; Hopke *et al.*, 2008; Cohen *et al.*, 2010; Fang *et al.*,

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2010). Results from inter-comparison studies of major Asian cities in most cases revealed high levels of PM₁₀ and PM_{2.5}, especially during dry season, and often they are in excess of USEPA 24-hrs and annual standards (Kim Oanh *et al.*, 2006; Ebihara *et al.*, 2008; Hopke *et al.*, 2008; Fang *et al.*, 2010). It is noted that, of these reported articles, only Ebihara *et al.* (2008) and Hopke *et al.* (2008) reported levels determined in Kuala Lumpur, Malaysia despite it being one of the major urban centres in Asia.

Research on air particulate pollution in Malaysia has been going on since late 90s, however until recently; results were mainly reported in the grey literature. For instance, Latif and Othman (1999) reported variation in 24-hrs total suspended particulate (TSP) concentrations for the industrial areas of Air Keruh, Malacca ($82 \pm 20 \mu\text{g}/\text{m}^3$) and Teluk Kalung, Terengganu ($68 \pm 15 \mu\text{g}/\text{m}^3$) and for the predominantly residential area of Bangi near to the Klang Valley ($102 \pm 35 \mu\text{g}/\text{m}^3$). The Malaysian Department of Environment (DOE), through its concessionaire company Alam Sekitar Malaysia (ASMA) Berhad, has been monitoring the PM₁₀ levels at 51 monitoring stations across Malaysia as part of the Malaysian Continuous Air Quality Monitoring (MCAQM) programme since 1996. Recently research has used this data to establish a clear link between monsoon seasonal variations with levels of PM₁₀ across the country (Juneng *et al.*, 2009; Md Yusof *et al.*, 2010). In addition, these data also confirmed the role of trans-boundary particulate matter in affecting the Malaysia air quality. Highest values were recorded during the worst haze experienced by Malaysia (August to September 1997) where all monitoring stations in Peninsular Malaysia were affected by the trans-boundary sources of biomass burning derived PM in Sumatra, Indonesia (Azmi *et al.*, 2010).

Chemical characterization and source apportionment studies on PM₁₀ (and below) are still sparse in Malaysia. The earliest work and most comprehensive chemical study to date is likely to be a one year monitoring programme (1997–1998) carried out by Hamzah *et al.* (2000). Their results showed that the fine fraction was the dominant fraction during the haze days being a factor of 4–5 times higher than non-haze days compared with a 2 fold increase in the coarse fraction. Based on the particulate elemental composition results of this study, they concluded that both natural and anthropogenic inputs contributed to the levels observed, with biomass burning source being dominant during the haze period. Assessment of elemental and water soluble ionic species content revealed the importance of vehicular emissions as a source followed by soil dust, most likely as a result of earthwork and land clearing activities in the vicinity.

Kuala Terengganu, the capital state of Terengganu, is one of the three major cities located on the east coast of West Malaysia (also commonly known as Peninsular Malaya) with an estimated population of over 360 000 occupying a land area of about 605 km². Located by the coast facing the southern part of South China Sea, this city is undergoing rapid development and urbanization. Air quality in the east coast is generally described as clean but in recent years some studies have indicated that whilst TSP (17.2–148 $\mu\text{g}/\text{m}^3$)

and PM₁₀ (52.9–134 $\mu\text{g}/\text{m}^3$) values were generally below the Recommended Malaysian Air Quality Guideline (RMAQG) for the respective fraction, some elemental composition of the TSP and PM₁₀ have clearly been affected by vehicular emission sources (Mohd Tahir *et al.*, 2008a, b). In addition, these studies have also demonstrated the importance of crustal and marine sources of particulate matters over Kuala Terengganu. A study on short-term temporal distribution of PM_{2.5} over Kuala Terengganu (Mohd Tahir and Khor, 2010) also revealed that this city can be affected by haze episodes with PM_{2.5} concentration exceeding the WHO 24h-guideline of 25 $\mu\text{g}/\text{m}^3$ during this period and their distribution is also influenced by monsoon seasonal variation. Long term studies on temporal distribution, chemical characterisation and source apportionment of particulate matter in PM_{2.5–10} (Coarse Particle, CP) and PM_{2.5} (Fine Particle, FP) simultaneously or independently in Malaysia is scarce and the DOE at present does not include FP parameter as part of her monitoring programme. In Malaysia, a guideline for FP fraction has yet to be developed and incorporated into the existing RMAQG, thus data generated from this study would provide valuable baseline information as Kuala Terengganu is strategically located in the east coast of the Peninsular. In addition, the results of the study will also help the state and federal government to plan their policy to reduce emission of particulate matter around the study area.

METHODS

Sampling Site and Method

The sampling campaign was initiated in Kuala Terengganu city centre (05°24'N, 103°06'E) from August 2006 until December 2007. A Gent PM₁₀ stacked filter unit (SFU) sampler provided by Malaysian Nuclear Agency was used for aerosol collection (Maenhaut *et al.*, 1993); the sampler unit was placed on Kuala Terengganu City Polytechnic (PKKT) building roof top 20 m above ground level (Fig. 1). The sampling inlet consists of a pre-impactor and two 47 mm Nucleopore polycarbonate filters with pore sizes of 8 (Apiezon coated) and 0.4 μm placed in series to collect coarse (PM_{2.5–10}) and fine (PM_{2.5}) particles, respectively. The air flow rate was maintained at between 14 and 16 L/min. Samples were collected twice a week and the collection time for each sample was over a period of 72 hrs. A total of 80 coarse and 80 fine samples were collected during this sampling campaign. The mass of particulate matters collected were determined gravimetrically using a micro-balance (Sartorius) to an accuracy of 0.1 mg. Prior to weighing, filters were pre-conditioned in a desiccator between 24–48 hrs to remove moisture. Immediately after mass determination, loaded samples will undergo chemical analyses. In cases where analyses were not immediately possible, sample filters were stored in the dark at ca. 4°C in a refrigerator to minimize the evaporation of volatile components. In general the filter storage time prior to chemical analysis did not exceed 2–3 days.

Chemical Analysis

Loaded filters were then divided into four equal portions;

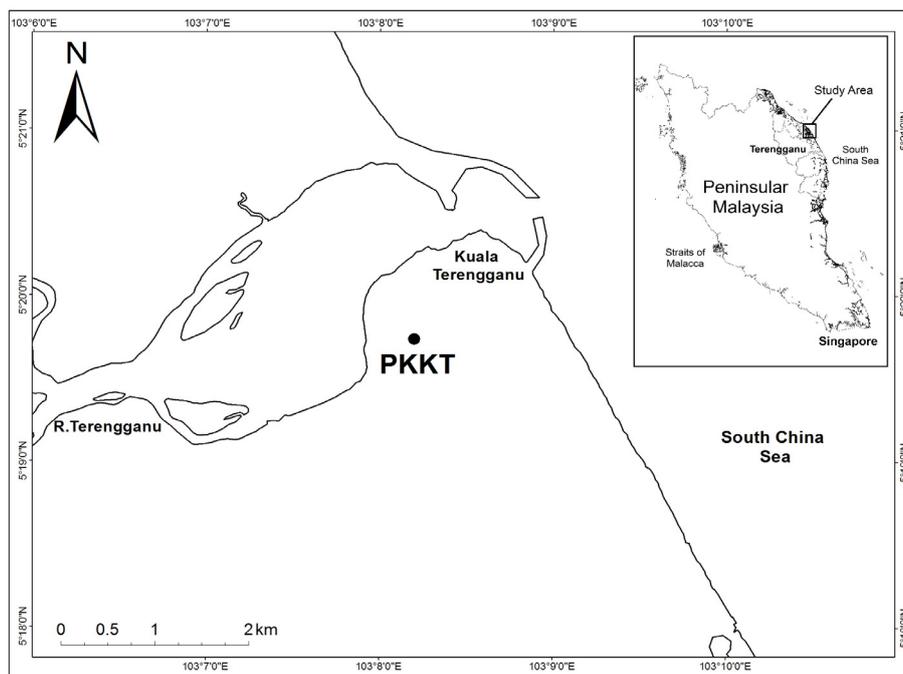


Fig. 1. Location of sampling site.

one portion was used for water soluble ionic species analysis (CEPA, 2007a) and the remaining portions were used for elemental analysis (CEPA, 2007b). In the case of elemental determination, the exposed filter was acid digested in a temperature programmed microwave oven followed by inductively coupled plasma-optical emission spectroscopy (ICP-OES) analysis (for major element determination) and inductively coupled plasma-mass spectroscopy (ICP-MS) analysis (for trace element determination). Water soluble ionic species extraction was carried out by ultrasonating the sample filter in de-ionised water followed by continuous shaking on a shaker for 12 hrs. The extracted solution was then subjected to ion chromatography for analysis of inorganic anions and cations. Standard reference material, (NIST SRM1648; urban particulate matter) and filter blanks were subjected to analytical protocols for quality control and assurance purposes. Recovery for metals and ions were in the range 84 to 95% and 82 to 118%, respectively.

RESULTS AND DISCUSSION

In this study, the concentration range of CP and FP fraction obtained varied from 1.17 to 28.45 $\mu\text{g}/\text{m}^3$ and from 3.82 to 35.58 $\mu\text{g}/\text{m}^3$, respectively. Fig. 2 shows the temporal variations of CP, FP and PM_{10} (defined as sum of coarse and fine particles, $\text{PM}_{10} = \text{CP} + \text{FP}$), as described by their monthly mean concentrations along seasonal trends throughout the sampling period. The variations in monthly concentration of CP, FP and PM_{10} as measured by the coefficient of variation was relatively large, ranging from 20.70 to 53.03%, 19.49 to 49.24% and 21.93 to 52.86%, respectively. A strong peak of particulate concentration was observed in October 2006 corresponding to a trans-boundary haze episode caused by forest fires in Sumatra

and Kalimantan, Indonesia (DOE, 2006). Result in Table 1 presents the descriptive statistics of the particulate mass concentration in Kuala Terengganu from August 2006 to December 2007. The overall mean particulate mass concentration of CP, FP and PM_{10} , were 10.41, 14.24 and 24.71 $\mu\text{g}/\text{m}^3$, respectively. The concentrations of all PM_{10} monitored during the study period were below the 24-hrs PM_{10} concentration suggested by Recommended Malaysian Air Quality Guideline (RMAQG). The result was in agreement with a study of the inhalable size fraction (PM_{10}) by Mohd Tahir *et al.* (2008) using a size selective high volume sampler over Kuala Terengganu. However, it is noteworthy that the PM_{10} concentrations observed in this study exceeded the annual RMAQG of 50 $\mu\text{g}/\text{m}^3$ three times within the study period; twice during the haze episode of October 2006 and once during April 2007. Currently, RMAQG does not provide value for $\text{PM}_{2.5}$. Comparison with the USEPA ambient air quality standard showed that none of the FP had exceeded the recommended 24-hrs standard value of 35 $\mu\text{g}/\text{m}^3$ but almost 33% of the FP observed had exceeded the USEPA annual guideline of 15 $\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$.

Table 2 shows the comparison of particulate matter obtained with those reported from selected cities in Asia. In addition, where possible, the percentage contributions of the FP fraction to PM_{10} were also included in Table 2 for comparison. It is acknowledged that these locations cover different areas ranging from industrial, traffic, urban, suburban dominated areas to rural areas thus making direct comparison invalid. Nonetheless, it is reasonable to conclude that the PM_{10} and fine particulate levels found in the present study were relatively lower than those reported in industrial, traffic and significant larger urban areas and comparable to those reported by Maenhaut *et al.* (2002) for Kalimantan

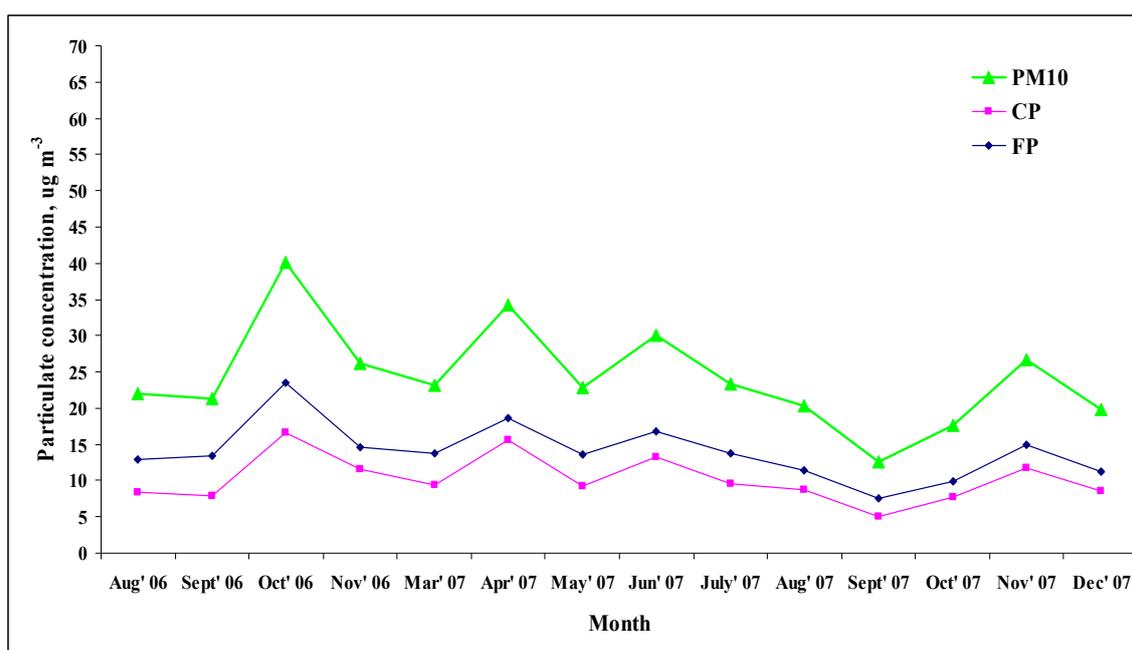


Fig. 2. Monthly mean particulate concentration.

Table 1. Particulate matter concentration (in $\mu\text{g}/\text{m}^3$; $n = 80$) in Kuala Terengganu.

	Mean	SD	Median	Ranges
CP	10.41	5.40	9.54	1.17–28.45
FP	14.24	6.60	13.18	3.82–35.58
PM ₁₀ *	24.71	11.63	22.71	5.21–63.48

CP: coarse particle; FP: fine particle; PM₁₀* = CP + FP; SD: standard deviation

and Sumatra, Indonesia and by Fang and Chang (2010) for Taiwan. Kuala Terengganu is a suburban area with no major industrial activities in the city and it is significantly less developed and populated than the major cities listed in the Table; the nearest industrial area is the Kerteh petrochemical industrial complex ca. 150 km south east of the city. It is interesting to note that the range and average values obtained in this study were much lower than those reported by Mohd Tahir *et al.* (2008). The differences could be attributed to methodological differences where sampling in the previous study was done at 1m above the ground using a high volume sampler, sampling frequency at a given site was limited ($n = 3$) and choice of sites within the city were source oriented (traffic related). The relationship of both CP and FP against the PM₁₀ concentration as illustrated in Fig. 3 shows that the CP and FP were significantly correlated with the PM₁₀, their relationship coefficients being $R^2 = 0.94$ and $R^2 = 0.95$ respectively. The higher slope of FP versus PM₁₀ (slope = 0.55) compared to CP versus PM₁₀ (slope = 0.44), indicates that the fine particles is more dominant component of PM₁₀ mass and has a significant influence on the observed variability of PM₁₀ at the site. The percentage of FP in PM₁₀ was fairly uniform, ranging from 50–77% with the average and standard deviation of $58 \pm 6\%$. Based on the overall mean particulate concentrations, FP was 1.4 times

the concentration of coarse particle. Samples from the site indicate a comparable FP percentage compared with those reported in selected industrial, traffic and high population cities listed in Table 2. A high percentage of FP in PM₁₀ has been suggested as being indicative of relatively high contributions from secondary particles and the combustion sources (Kim Oanh *et al.*, 2006). The variation of all particulate concentration in this study was high with the coefficients of variation (CV) of 28.4%, 35.5% and 63.4% for CP, FP and PM₁₀, respectively, which suggests that the local meteorological variables as well as the regional air pollution sources are important in determining air quality at the site. Correlation analysis between CP and FP showed that they were significantly correlated with $r = 0.89$ ($p < 0.01$). This high correlation coefficient between these two particulate fractions suggests that they could originate from similar particulate sources and also implies that the processes of generation of these particulate fractions are linked.

Source of Air Particulate Matter

Figs. 4, 5 and 6 show the mean concentration of water soluble ionic species (WSIS), major elements and minor elements found in the air particulate fractions, respectively. WSIS measured in this study accounted ca. 30% and ca. 26% of the coarse and fine particle mass, respectively. Sodium, chloride, sulphate and nitrate were the four major ions present in the CP fraction which accounted for 80.1% of the total soluble ion measured in this fraction. On the other hand, sulphate, ammonium, potassium and nitrate ions are the four major species in the FP fraction which accounted for 92.3% of the total soluble ionic species measured in the FP.

Elements measured accounted for ca. 24% and ca. 6% of the total CP and FP mass, respectively. Al, Ca, Fe and Na dominated the identifiable elemental component in CP and

Table 2. Comparison of PM₁₀, fine and coarse particulate matter ($\mu\text{g}/\text{m}^3$) with other studies in Asia.

Reference	Location	Character	PM ₁₀	Fine	Coarse	%FP/PM ₁₀
This study	Kuala Terengganu, Malaysia	Coastal, sub-urban	5.21–63.48 24.71 ± 11.63*	3.82–35.58 14.3 ± 6.5*	1.17–28.45 10.4 ± 5.4*	58
Hamzah et al. (2000)	Kuala Lumpur	Urban (haze days: Sept 15–26, 1997) Non-haze (1997–98)	225 ± 99* 55.4 ± 30.0*	174 ± 95* 30.9 ± 14.4*	50 ± 26* 24.5 ± 21.1*	77 56
Mohd Tahir and Khor (2010)	Kuala Terengganu, Malaysia	Coastal, sub-urban	9.48*			
Mohd Tahir et al. (2008)	Kuala Terengganu, Malaysia	Coastal, sub-urban	52.9–134; 75.1*			
Norela et al. (2010)	Kuala Lumpur, Malaysia	Urban	4–20			
Azmi et al. (2010)	Klang Valley, Malaysia Jerantut, Malaysia	Urban, Industrial Rural	22–218; 56.5* 17–152; 38.6*			
Balasubramaniam et al. (2003)	Singapore	Coastal, industrial, urban	27.2*			
Maenhaut et al. (2002)	Sumatra, Indonesia Kalimantan, Indonesia	Mountain site, remote Sea level site, rural	9.3* 21.4*	4.5* 9.4*	4.8* 12*	48 44
Hien et al. (2001)	Ho Chi Minh, Vietnam	Urban, residential, traffic	16	16	32*	
Chuersuwan et al. (2008)	Bangkok, Thailand	Urban Residential Low impact urban	108.1* 61.6* 57.6*	69.0* 41.2* 37.9*		64 67 66
Hopke et al. (2008)	China	Urban Sub-urban		4.6–96.2; 28.04* 7.1–179.5; 42.14*	5.7–196.1; 58.3* 9.1–387; 109*	
Ho et al. (2006)	Manila, Philippines	Residential		3–51; 27.2*	0–157; 22.8*	
Fang and Chang (2010)	Hong Kong	Urban, traffic	43*			
Lim et al. (2010)	Taiwan Korea	Suburban, rural, traffic Industrial Residential	53* 87.2* 73.9*	29.3*		55

*: mean value

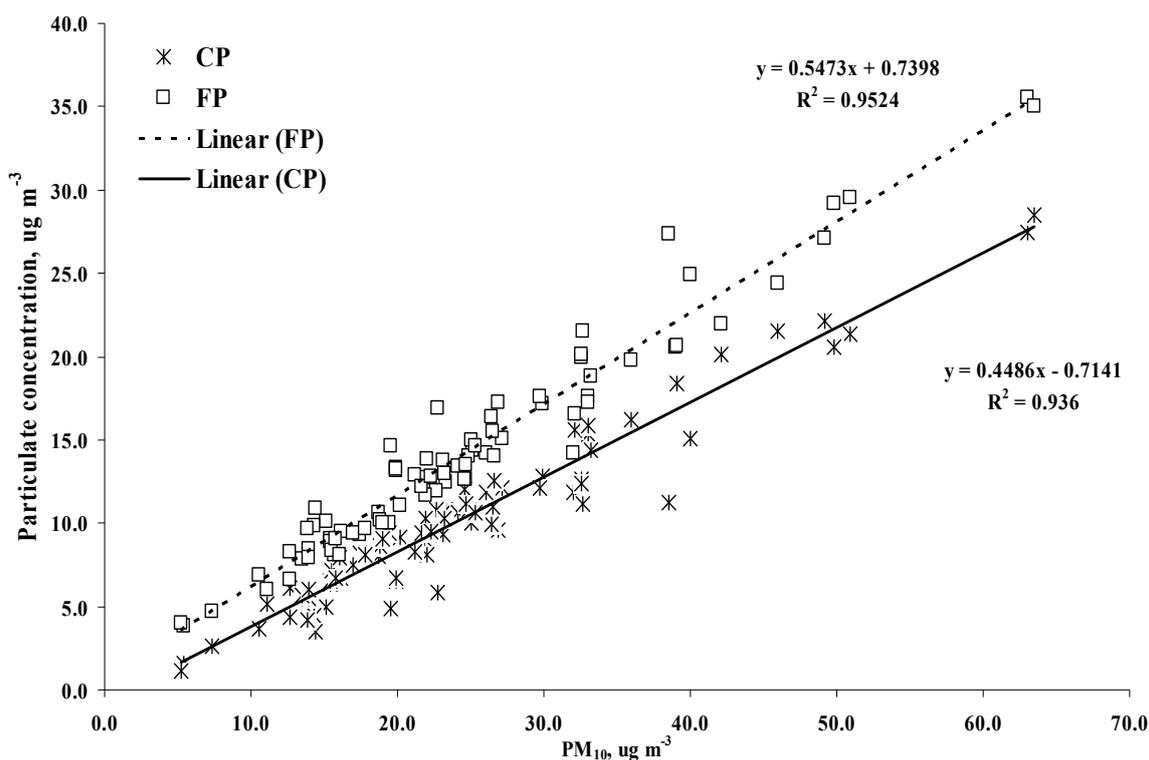


Fig. 3. Relationship of CP and FP versus PM_{10} .

represented ca. 83% of the total measured elemental concentration of the CP fraction. On the other hand Al, K, Fe, Mg and Zn dominated the elemental component in FP and represented ca. 80% of the total measured elemental concentration of the FP fraction.

On average, measured chemical species in this study accounted for ca. 54% of the coarse particle fraction and ca. 32% of the fine particle. Na^+ , SO_4^{2-} , Cl^- , NO_3^- , Al, Ca, Fe, K and Na were mainly in coarse particle size range and accounted for ca. 52% of the coarse particle mass, while SO_4^{2-} , NH_4^+ , K^+ , Al, Ca, K and Na were mainly in fine particle size range and accounted for ca. 28% of the fine particle mass. It is postulated that the remaining major composition would probably consist of most organic or carbonaceous materials. In this study only selected elements and water soluble inorganic ions were measured whilst other chemical constituents such as Water Soluble Organic Carbons (WSOC), Black carbon (BC), Elemental Carbon (EC) and other organic carbon (OC) were not measured due to lack of instrumentation; these constituents could account over 40–70% of the gravimetric mass fraction (Hamzah et al., 2000; Balasubramanian et al., 2003). Hamzah et al. (2000) reported mass closure of 15–40% for FP and 50–75% for CP despite them measuring the elemental carbon (EC) but leaving out sulphur or sulphate and organic carbon measurements. Balasubramanian et al. (2003), on the hand, were able to obtain a good mass balance agreement between the sum of the determined groups of aerosol components (which include WSIS, WSOC, EC, OC and trace elements) and the gravimetrically determined mass; this study demonstrated the importance of organic or carbonaceous constituents to obtain a mass balance in chemical analysis of APM.

Enrichment Factor

Elements in aerosols are derived from multitude of sources and the degree to which these elements in the aerosols are enriched or depleted, relative to a specific source, can be assessed using an Enrichment Factor Analysis. For crustal source, Al is normally used as the source indicator element and the earth's crust as the source material (Chester et al., 1997). The abundance of elements in the upper continental crust was taken from Wedepohl (1995). Typically, the calculation of a normalized enrichment factor for a given element is represented by Eq. (1).

$$EF_{\text{crust}} = (E_x/Al)_{\text{sample}} / (E_x/Al)_{\text{crust}} \quad (1)$$

where E_x is the median concentration of the element of interest. $(E_x/Al)_{\text{sample}}$ is the concentration ratio of element X to Al in aerosol sample and $(E_x/Al)_{\text{crust}}$ is the concentration ratio of X to Al in the crustal material. By convention, an $EF < 10$ is taken as an indication that an element in an aerosol has a dominant crustal source whereas an $EF > 10$ is considered as indicative of significant contribution from non-crustal source.

Fig. 7 shows the median EF_{crust} values of individual elements in both coarse and fine size fraction. One distinct feature observed is that elements were more enriched in the fine than the coarse fraction. Fig. 6 also shows that Zn, Cd, Ni, Cr, Pb and Cu were enriched in both coarse and fine particles. In most cases, these elements were related to anthropogenic emissions, such as combustion of fossil fuel, automobile exhaust emissions and other traffic related contributions (Chan et al., 1997; Lim et al., 2010). In contrast, EF_{crust} values of Ca, Fe, K, Mg, Na, Mn and Co in both

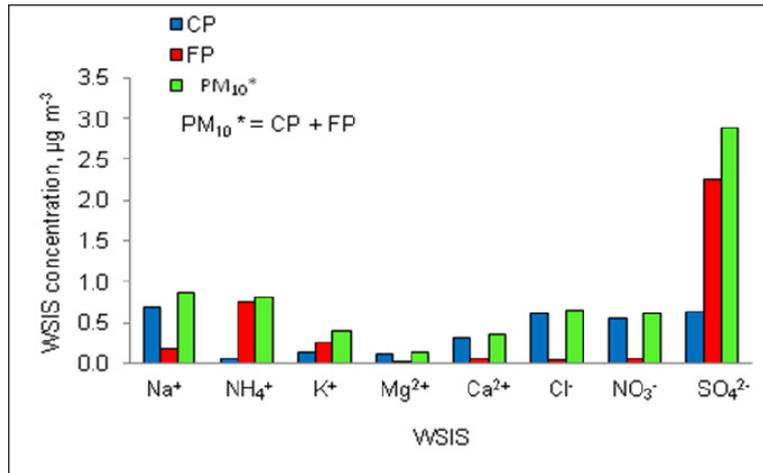


Fig. 4. Mean concentration of water soluble ionic species (WSIS).

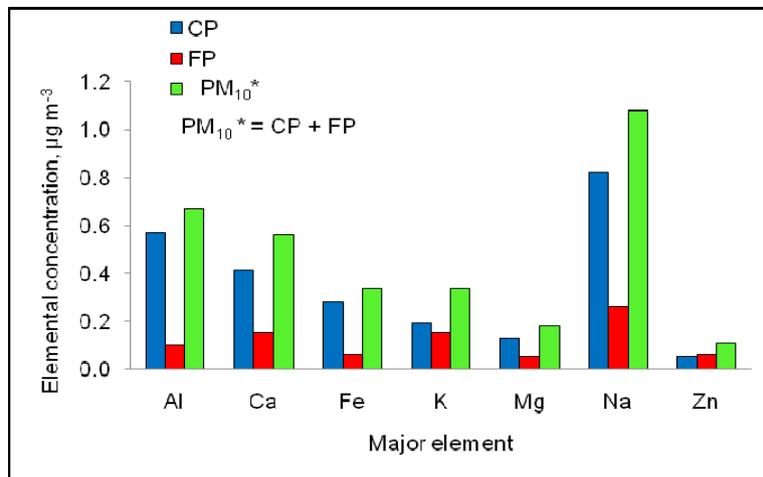


Fig. 5. Mean concentration of major elements.

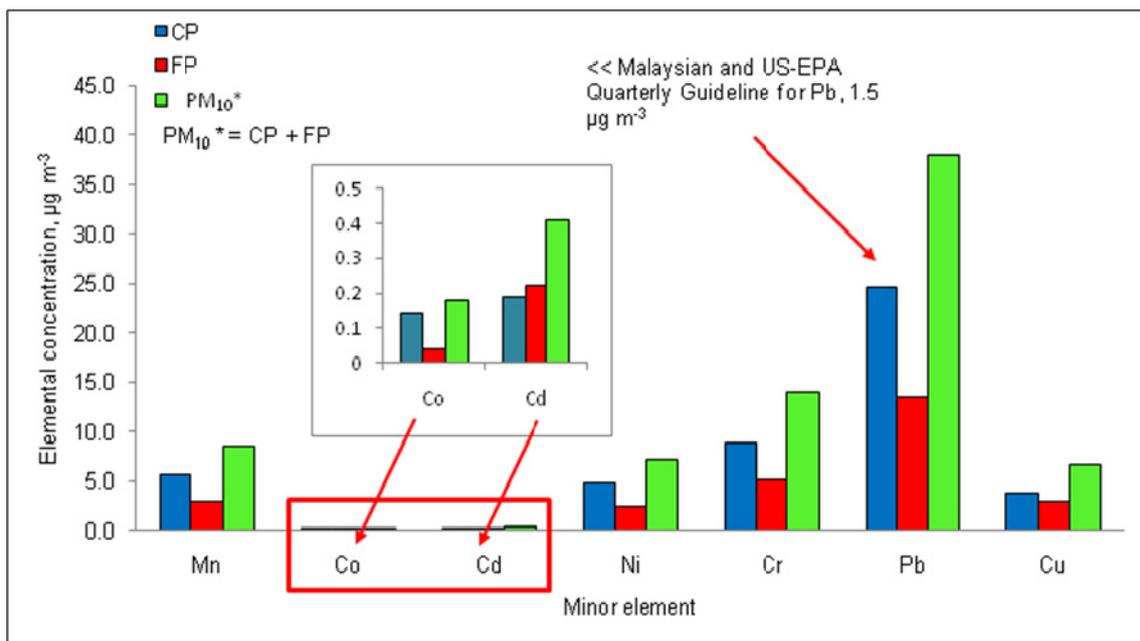


Fig. 6. Mean concentration of minor elements.

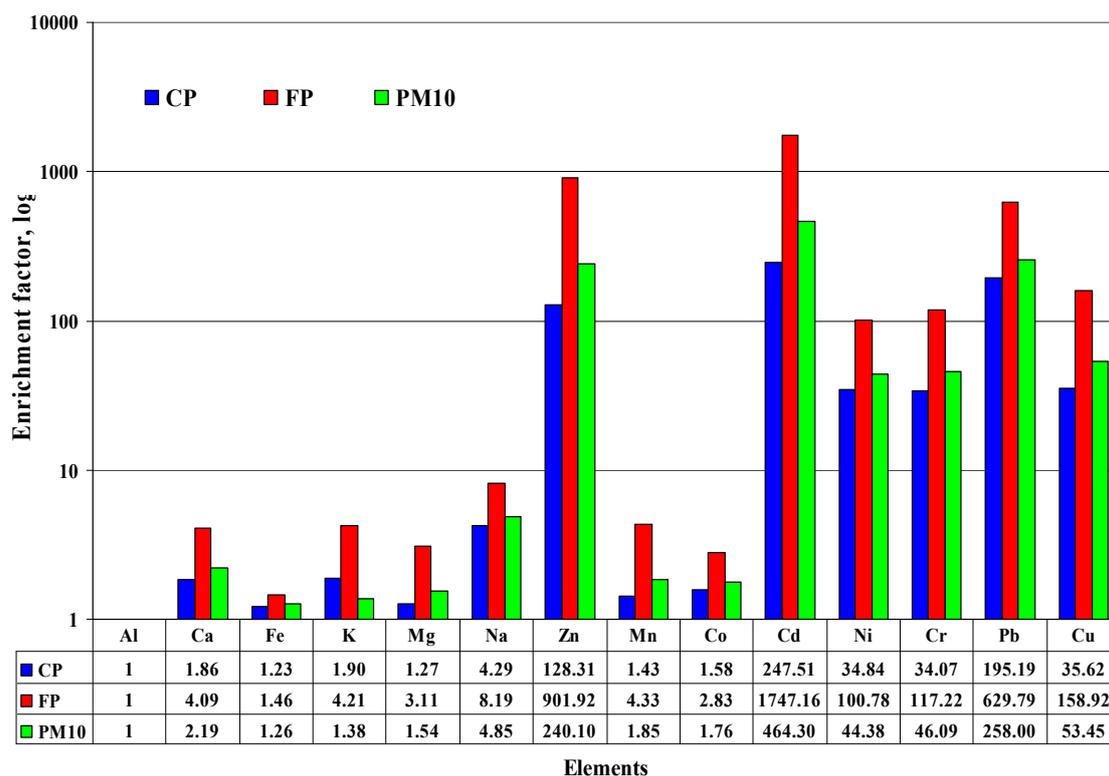


Fig. 7. Enrichment factor of elements in CP, FP and PM₁₀ (CP + FP).

coarse and fine particles were less than 10, suggesting the dominance of natural origin, consistent with their presence primarily in coarse particles. However, these elements were slightly enriched in the fine particle fraction relative to the coarse suggesting possible additional association with input from non-local crustal origin. These findings illustrate that the sources of these elements in the coarse and fine particles may be different and involve different mechanisms in their production. Review of the literature suggests that anthropogenic input of Ca could possibly be from the cement industry, Fe and Mn were primarily from industrial metallurgical process and Co could be from smelters (Cohen *et al.*, 2010; Hieu and Lee, 2010). However, in the absence of these industrial activities in study area, it is hypothesized that the enrichment of crustal component in fine particles relative to the CP could be associated with the long-range transport from their sources (Yongjie *et al.*, 2009). In order to verify this hypothesis, an attempt was made to evaluate the non-crustal contribution to these elements using a method suggested by Wang *et al.* (2006) where it is assumed that the total mass of an element associated with aerosol particles is a sum of contributions from crustal and non-crustal origins as described below:

$$[E_x]_{Air} = [E_x]_{Crust} + [E_x]_{Non-crustal} \quad (2)$$

where the subscripts Crust and Non-crustal are designated as crustal and non-crustal fractions. Since only 11% of Al concentration was detected in fine particles in this study, it was assumed that Al was from natural sources only, while anthropogenic emissions accounted for a negligible fraction,

i.e., $[Al]_{Air} = [Al]_{Crust}$, Eq. (2) can be rearranged as follows:

$$[E_x]_{Non-crustal} = [E_x]_{air} - [Al]_{Air} \times [E_x/Al]_{Crust} \quad (3)$$

Results from the calculation revealed that percentage contribution of non-crustal sources for Ca, Fe, K, Mg, Na, Mn and Co in fine particles ranged between 28–48% on elements (K > Co > Mn > Mg > Ca > Fe > Na) suggesting that crustal source is still the dominant contributor in the fine particle fraction.

Principal Component Analysis

Principle Component Analysis (PCA) was applied to the coarse and fine particulate compositions. Tables 3 and 4 present the factor loading, eigenvalues and percentage of total variance obtained for CP and FP, respectively. There were six main chemical profile sources contributing to fine particles identified (corresponding to eigenvalues greater than 1 Table 4); these profiles were mineral aerosol, vehicle exhaust, marine aerosol, traffic aerosol, secondary aerosol and biomass burning. These sources accounted for 73% of the total variance in the PCA. For coarse particles, the principle component accounted for 80% of the total variance of the original data which had similar source profiles with fine particles but with slight variation in the order of importance.

Soil

In both fraction, the first principle component (PC) presented high factor loading for Al, Ca, Fe, K, Mg, Na, Mn, Co, Na⁺, Ca²⁺, K⁺, Mg²⁺ and accounted 32% and 34% of total variance in coarse and fine particles, respectively.

Table 3. PCA of CP speciation data.

	PC1	PC2	PC3	PC4	PC5	PC6
	Soil	Sea	Vehicle exhaust	Secondary aerosol	Traffic	Biomass burning
log Al	-0.83	-0.13	0.22	-0.22	0.05	0.08
log Ca	-0.55	-0.25	-0.03	0.17	-0.26	0.15
log Fe	-0.70	-0.30	0.23	-0.33	0.14	0.05
log K	-0.82	-0.22	-0.18	0.15	0.10	-0.59
log Mg	-0.48	-0.53	-0.13	0.23	-0.10	-0.03
log Na	-0.53	-0.73	-0.23	0.16	-0.03	-0.28
log Mn	-0.81	0.00	0.38	-0.18	0.04	-0.21
log Co	-0.77	0.06	0.35	0.03	0.04	-0.32
log Zn	-0.11	0.19	-0.40	0.35	-0.71	0.08
log Cd	-0.47	-0.04	-0.67	0.15	-0.54	-0.09
log Ni	-0.38	0.16	-0.78	-0.09	-0.51	-0.13
log Cr	-0.05	0.07	-0.74	-0.02	-0.60	-0.25
log Pb	-0.27	0.09	-0.57	0.15	-0.67	-0.32
log Cu	-0.37	-0.09	-0.57	0.15	-0.77	-0.06
log Na ⁺	-0.57	-0.74	-0.04	-0.04	-0.02	-0.05
log NH ₄ ⁺	-0.23	0.18	0.26	-0.69	0.25	-0.53
log K ⁺	-0.49	0.24	0.06	-0.07	0.21	-0.69
log Mg ²⁺	-0.48	-0.70	-0.20	-0.10	-0.04	-0.18
log Ca ²⁺	-0.66	0.10	0.13	-0.23	-0.25	0.04
log Cl ⁻	-0.26	-0.82	-0.12	0.05	0.05	0.04
log NO ₃ ²⁻	-0.24	0.23	-0.55	-0.78	-0.23	0.21
log SO ₄ ²⁻	-0.31	-0.41	-0.65	-0.73	-0.29	0.29
Eigenvalue	7.47	4.20	2.93	1.44	1.21	1.18
Cumulative eigenvalue	7.47	11.67	14.59	16.04	17.25	18.43
% total of variance	32.46	18.27	12.72	6.30	5.27	5.11
Cumulative, %	32.46	50.73	63.45	69.75	75.02	80.13

This pattern of elements indicates their origin from soil as it contains typical elements of crustal origin (Poh, 2008; Yongjie *et al.*, 2009). Since Kuala Terengganu is a coastal city, it is not surprising that its soils contain relatively high concentration of elements like Na, Ca, K and Mg attributed to accumulation of marine aerosols through atmospheric deposition into the soils (Poh, 2008). This finding is consistent with that EF analysis.

Marine Aerosols

PC 2 in the coarse fraction (18.3% of total variance) and PC3 in the fine fraction (9.1% of total variance) have been associated with the marine aerosol since these are characterized by high loading on typical marine composition such as Na, Na⁺, Cl⁻ and Mg²⁺ (Seinfeld and Pandis, 2006). Higher concentration of Na⁺ and Cl⁻ was observed in the CP. It is believed the sea salt concentration was high at the measured site as a result of the higher and more persistent on-shore winds which create high sea water droplets and marine aerosols. A robust ratio of Cl⁻ to Na⁺ was obtained for CP (1.3) compared to FP (0.02); the lower ratio in the latter fraction has been attributed to chloride loss as a result of reactions between acidic species, mainly HNO₃ and H₂SO₄, with NaCl in sea salt particles (Khan *et al.*, 2010 and references therein). Evaluation of chloride loss showed percent loss ranging between 0.2–91% (mean 59.8%) in CP and 25.9–99.6% (mean 90.3%) in FP respectively, suggesting the percentage of chloride loss from sea salt particles

decreased with increasing of particle size. This finding is in agreement with Kerminen *et al.* (2000) in Antarctic atmosphere where the average of chloride loss was > 90% for submicron particles (< 3 μm) and slightly above 50% for particles larger than about 3 μm. Interestingly, SO₄²⁻ in CP was also found to be well correlated with this marine component. However, evaluation of non sea salt sulphate (nss-SO₄²⁻) revealed sea salt SO₄²⁻ only accounted for ca. 28.4% of total mean sulphate in CP compared to ca. 2% in FP.

Vehicle Exhaust

NO₃⁻, SO₄²⁻, Zn, Cd, Ni, Cr, Pb and Cu present a strong correlation with PC3, in the coarse particle fraction (12.7% of total variance), and PC2 in the fine particle fraction (12.1% of total variance), respectively. The possible source of NO₃⁻, SO₄²⁻, Zn, Cd, Ni, Cr, Pb and Cu in both fractions was vehicle exhaust emissions (Pio *et al.*, 1996; Samara *et al.*, 2003; Ozaki *et al.*, 2004; Amorim *et al.*, 2007).

Secondary Aerosols

In the both fractions, PC4 represents the secondary aerosol production (6.3 and 8.1% of total variance for CP and FP fractions, respectively). This PC presents high factor loadings for SO₄²⁻, NH₄⁺ and NO₃⁻. SO₄²⁻ and NH₄⁺ were derived from the gas to particle conversion process resulting from SO₂ oxidation and NH₃ neutralization and are the main secondary aerosol components defining particulate matter

Table 4. PCA of FP speciation data.

	PC1	PC2	PC3	PC4	PC5	PC6
	Soil	Vehicle exhaust	Sea	Secondary aerosol	Traffic	Biomass burning
log Al	-0.69	-0.29	-0.19	0.42	0.08	-0.04
log Ca	-0.66	-0.12	-0.24	-0.03	-0.29	-0.13
log Fe	-0.56	-0.20	-0.33	0.33	0.33	-0.05
log K	-0.50	-0.15	0.17	0.25	-0.12	-0.58
log Mg	-0.72	0.10	-0.41	-0.06	-0.34	-0.06
log Na	-0.43	-0.19	-0.70	0.26	-0.16	0.12
log Mn	-0.43	-0.28	0.27	-0.27	0.10	0.34
log Co	-0.57	-0.23	-0.16	-0.29	0.19	0.04
log Zn	-0.16	-0.47	-0.02	-0.26	-0.76	0.09
log Cd	-0.41	-0.74	-0.01	-0.14	-0.47	0.03
log Ni	-0.13	-0.63	0.13	-0.38	-0.58	-0.07
log Cr	0.21	-0.45	-0.17	-0.11	-0.44	-0.21
log Pb	-0.52	0.15	-0.08	-0.26	-0.54	-0.18
log Cu	-0.22	-0.30	0.16	-0.19	-0.72	-0.20
log Na ⁺	-0.39	0.27	-0.44	0.10	0.10	0.12
log NH ₄ ⁺	-0.29	0.20	0.26	-0.45	-0.09	-0.49
log K ⁺	-0.47	-0.01	-0.54	0.30	0.02	-0.67
log Mg ²⁺	-0.59	0.25	-0.39	0.09	-0.01	0.18
log Ca ²⁺	-0.65	0.26	0.18	0.02	0.11	-0.14
log Cl ⁻	-0.13	-0.10	0.00	-0.43	0.15	0.26
log NO ₃ ²⁻	-0.56	0.31	0.08	-0.63	-0.26	0.05
log SO ₄ ²⁻	-0.11	-0.41	0.24	-0.66	-0.34	0.15
Eigenvalue	7.77	2.77	2.08	1.86	1.25	1.13
Cumulative eigenvalue	7.77	10.54	12.63	14.48	15.75	16.87
% total of variance	33.78	12.06	9.05	8.09	5.48	4.89
Cumulative, %	33.78	45.84	54.89	62.99	68.47	73.36

composition in air (Almeida *et al.*, 2005). In the fine fraction, these ions were strongly associated with each other indicating that SO₄²⁻ was mostly present as ammonium sulphate.

Traffic Aerosols (Non-engine Combustion emission)

Zn, Cd, Ni, Cr, Pb and Cu were correlated in PC5 with 5.3% and 5.9% of total variance for CP and FP fraction, respectively. These elements are usually associated with various sources namely coal combustion, vehicular emission and incineration activities (Morawska and Zhang, 2002; Manno *et al.*, 2006; Colbeck, 2008). However, owing to the absence of industrial sources in the study area, non combustion engine emission sources such as tire, clutch and brake wear, as opposed to non-vehicular combustion processes, was probably the source of these elements.

Biomass Burning

In both fractions, PC6 (5.1% and 4.5% of total variance for CP and FP fractions, respectively) was dominantly loaded by K, K⁺ and NH₄⁺. These species were tracer of biomass burning (Chan *et al.*, 1997; Khan *et al.*, 2010). Open burning of garden wastes and use of wood as fuel is still a common practice in this city. Consequently, contribution of biomass burning to air particulate matter concentration from biomass burning could be substantial source. In addition, K and K⁺ in this study could also be attributed to the use of palm fiber and shell waste (as boiler fuel) from regional palm oil mills (Subramaniam *et al.*, 2008). Currently, there are

more than 250 palm oil mill plants operating in Peninsular Malaysia with 14 in the state of Terengganu (MPOB, 2007).

CONCLUSIONS

The mean concentration of coarse and fine particle during the study period was 10.54 ± 5.42 µg/m³ (n = 80) and 14.24 ± 6.60 µg/m³ (n = 80), respectively. The average concentration of fine particle was lower compared to the US National Ambient Air Quality Standard (24-hrs average of 35 µg/m³). On average, measured chemical species accounted for ca. 54% of the coarse particle fraction with Na⁺, SO₄²⁻, Cl⁻, NO₃⁻, Al, Ca, Fe, K and Na dominance representing ca. 52% of the coarse particle mass. On the other hand, measured chemical species accounted ca. 32% of the fine particle with SO₄²⁻, NH₄⁺, K⁺, Al, Ca, K and Na being the more dominant species representing ca. 28% of the fine particle mass. Crustal enrichment factor (cEF) analysis showed that elements in the FP were generally more enriched than the corresponding CP. Of the elements measured, Zn, Cd, Ni, Cr, Pb and Cu were found to be significantly enriched in both particle fractions indicative of non-crustal input i.e. of anthropogenic origin. Principle Component Analysis carried out on the elemental and water soluble ionic species content of these fractions showed that soil dust, marine aerosol, vehicle exhaust, secondary aerosol, traffic aerosol (non-engine combustion) and biomass burning were the main sources of these particulate fractions.

ACKNOWLEDGEMENTS

The authors wish to thank Malaysian Nuclear Agency for the loan of Gent Stack sampler and funding (TE67913) which enabled the commencement of this work. The financial support from the Ministry of Higher Education, Malaysia (FRGS grant no. 59063) contributed to the completion of the project. A scholarship to FTS by UMT's postgraduate financial scheme is also acknowledged. The laboratory staff of Department of Chemical Sciences, UMT is acknowledged for their technical support with ICP-OES and IC. Thanks also to Dr. Keith Weston (Cefas, U.K.) for assistance with the proof reading.

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Received for review, August 17, 2012

Accepted, December 10, 2012