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Meteorological-gaseous influences on seasonal $PM_{2.5}$ variability in the Klang Valley urban-industrial environment

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Abstract

This study attempts to investigate the fine particulate matter ($PM_{2.5}$) variability in the Klang Valley urban-industrial environment. In total, 94 daily $PM_{2.5}$ samples were collected during a one-year campaign from August 2011 to July 2012, covering all four seasons. The samples were analysed for various inorganic components and black carbon. The chemical compositions were statistically analysed and the aerosol pattern was characterised using descriptive analysis, correlation matrices, enrichment factors (EF), stoichiometric analysis and chemical mass closure (CMC). For source apportionment purposes, a combination of positive matrix factorisation (PMF) and multi-linear regression (MLR) was employed. Further, meteorological-gaseous parameters were incorporated into each analysis for improved assessment. The results showed that $PM_{2.5}$ mass averaged at $28 \pm 18\,\mu g\,m^{-3}$, 2.8 fold higher than the World Health Organisation (WHO) annual guideline. On a daily basis, the $PM_{2.5}$ mass ranged between 6 and $118\,\mu g\,m^{-3}$ with 43 % exceedance of the daily WHO guideline. The North-East monsoon (NE) was the only season with < 50 % sample exceedance of the daily WHO guideline. On an annual scale, $PM_{2.5}$ mass correlated positively with temperature (T) and wind speed (WS) but negatively with relative humidity (RH). With the exception of NO_x, the gases analysed (CO, NO_2, NO and SO_2) were found to significantly influence the $PM_{2.5}$ mass. Seasonal variability unexpectedly showed that rainfall, WS and wind direction (WD) did not significantly correlate with $PM_{2.5}$ mass. Further analysis on the $PM_{2.5}$ / PM_{10}, $PM_{2.5}$ / TSP and PM_{10} / TSP ratios reveal that meteorological parameters only greatly influenced the coarse particles (PM > 2.5µm) and less so the fine particles at the site. Chemical composition showed that both primary and secondary pollutants of $PM_{2.5}$ are equally important, albeit with seasonal variability. The CMC components identified were: black carbon (BC) > secondary inorganic aerosols (SIA) > dust > trace elements (TE) > sea salt > K^+. The EF analysis distinguished two groups of trace elements: those with anthropogenic sources (Pb, Se, Zn, Cd, As, Bi, Ba, Cu, Rb, V and Ni) and those with a crustal source (Sr, Mn, Co and Li). The five identified

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factors resulting from PMF 5.0 were: (1) combustion of engine oil; (2) mineral dust; (3) mixed SIA and biomass burning; (4) mixed traffic and industrial; and (5) sea salt. Each of these sources had an annual mean contribution of 17, 14, 42, 10 and 17 %, respectively. The dominance of each identified source largely varied with changing season and a few factors were in agreement with the CMC, EF and stoichiometric analysis, accordingly. In relation to meteorological-gaseous parameters, $PM_{2.5}$ sources were influenced by different parameters during different seasons. In addition, two air pollution episodes (HAZE) revealed the influence of local and/or regional sources. Overall, our study clearly suggests that the chemical constituents and sources of $PM_{2.5}$ were greatly influenced and characterised by meteorological and gaseous parameters which largely vary with season.

1 Introduction

Airborne particulate matter (PM) significantly impacts global climate (Jacobson, 2002), causing visibility degradation in both urban and pristine environments (Diederen et al., 1985; Doyle and Dorling, 2002; Watson, 2002; Chang et al., 2009; Hyslop, 2009) and accelerates material decay (Grossi and Brimblecombe, 2002). Different sizes of PM have been found to have varying toxicities impacting human health (Schwartz et al., 1996; Katsouyanni et al., 1997; Pope III, 2000; Ruuskanen et al., 2001; Eatough et al., 2003; Halonen, 2009). Composed of both stable and semi-volatile compounds, fine particles appear to do more harm to human health than coarse particles (Dockery et al., 1993; Schwartz et al., 1996; Laden et al., 2000; Lanki et al., 2006; Pope III and Dockery, 2006).

The fraction and composition variability of fine particles ($PM_{2.5;}$ $dp < 2.5\,\mu m$) are strongly influenced by seasonal meteorological factors, gaseous parameters and location. Megaritis et al. (2014) had shown that $PM_{2.5}$ in Europe appears to be more sensitive to temperature changes compared to other meteorological and gaseous parameters in all seasons. Aside from meteorological and gaseous pollutants, seasonal

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changes and background of an area also influenced the $PM_{2.5}$ chemical variability (Tai et al., 2010, 2012). Seasonal variation of $PM_{2.5}$ mass and its chemical composition for the Asian region has been widely reported. Balasubramanian et al. (2003) reported that Singapore $PM_{2.5}$ mass temporal variability was influenced by a number of factors including changes in emission strength, wind direction (WD) and other meteorological parameters while their chemical mass closure (CMC) at times was significantly attributed by Indonesian forest fires compared to local traffic and industrial emissions. Ye et al. (2003) reported varied CMC elements for Shanghai seasons where significant changes in the $PM_{2.5}$ mass were observed with changing season. Meanwhile, sources of $PM_{2.5}$ in Beijing showed distinct seasonal trends (Zheng et al., 2005) and Indian $PM_{2.5}$ sources were observed to have considerable seasonal and weekday/weekend variations (Srimuruganandam and Shiva Nagendra, 2012b). A study by Louie et al. (2005) on $PM_{2.5}$ chemical compositions showed variations between different locations in Hong Kong where elevated concentrations of a source marker species at a site explained a higher influence of that source. Similar observations were also evident for Indonesia where source apportionment analysis on the elemental composition of PM revealed different numbers of factors for urban and suburban areas (Santoso et al., 2008).

$PM_{2.5}$ in the atmosphere consists of primary and secondary pollutants including volatile, non-volatile and semi-volatile components which originate from various sources (Eatough et al., 2006). Source apportionment (SA) is an approach that aims to identify and quantify the various sources of air pollutants (Hopke and Song, 1997; Watson et al., 2002; Wagstrom and Pandis, 2011). The most common method is receptor modelling which measures atmospheric concentrations of chemically-speciated particles to infer the sources responsible for their emission or the pathways of formation of secondary particles (Viana et al., 2008). The method starts by collecting and measuring ambient PM at a receptor (location), and works backwards to determine the sources. The goal of receptor models is to solve the chemical mass balance between measured species concentrations and source profiles. One of the models used to solve

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the chemical mixture is positive matrix factorisation (PMF), first developed by Paatero and Tapper (1993). Subsequently, numerous other studies have employed this method into their $PM_{2.5}$ receptor modelling studies. For example, Begum et al. (2004) and Santoso et al. (2008) have successfully applied PMF version 2 (PMF2) on inorganic and black carbon datasets to lead to source identification for $PM_{2.5}$ in Bangladesh and Indonesia, respectively. A study by Rahman et al. (2011) also used similar chemical composition for Klang Valley $PM_{2.5}$ SA analysis but using PMF version 3 (PMF3). Srimuruganandam and Shiva Nagendra (2012b) has made an evaluation of $PM_{2.5}$ sources for Chennai city, India by using only inorganic (elemental) composition. A study by Zhang et al. (2013) has successfully discussed the seasonal perspective of $PM_{2.5}$ sources in Beijing, China using PMF3 on inorganic and organic datasets. The latest version of PMF, i.e. PMF5, was effectively applied by Khan et al. (2015b) to their polycyclic aromatic hydrocarbons (PAHs) dataset to characterise the $PM_{2.5}$ for the semi-urban area of Bangi, Malaysia. One of the current trends of SA is to apply more than one receptor model, a trend set by a number of countries i.e. Belgium, Germany, Portugal and Spain (Viana et al., 2008). This study also reports that the most frequent combinations used for SA are principal component analysis (PCA)-cluster analysis (CA), PCA-Lenschow, PCA-chemical mass balance (CMB), PCA-back-trajectory analysis, PMF-UNMIX-multilinear engine (ME), and CMB-mass balance.

A study by Reid et al. (2013) discussed in detail how the Southeast Asian (SEA) region holds a complex relationship between geographic, socio-economic, meteorological, and aerosol microphysical factors. The review emphasised timing and location of sampling in getting the actual condition of the aerosol system. Taking this into consideration, we conducted a one-year assessment covering all four seasons (including haze events) to investigate the $PM_{2.5}$ variability in the Klang Valley. The samples were subjected to chemical measurements of inorganic matter (IM) compositions and black carbon (BC). We identified and apportioned the sources to $PM_{2.5}$ mass by employing CMC construction and the PMF-MLR model in conjunction with trajectory clusters. All variables of $PM_{2.5}$ mass, their chemical compositions identified, as well as the sources

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predicted, were further analysed using correlation matrices with the meteorological-gaseous pollutants for comprehensive assessment.

2 Material and methods

2.1 Sampling site description

The sampling took place on the rooftop of the Malaysian Meteorological Department (MET) located in the city of Petaling Jaya (MET PJ; 3°06′09.2″ N 101° 38′41.0″ E), about 100 m above the sea level (Fig. 1). This site was chosen to represent the region of Klang Valley on the western side of Peninsular Malaysia. The Klang Valley area is the heartland of industry and commerce in Malaysia and is densely populated (Azmi et al., 2010). MET PJ is 10 km west of Kuala Lumpur, the capital city of Malaysia. This sampling site is part of the principal station for MET and in addition, the site is also one of the Global Atmosphere Watch (GAW) Regional Station representing the tropical region of the World Meteorological Organisation WMO-GAW network. This site is regarded as being representative of urban-industrial conditions, categorised according to criteria proposed by the Malaysia's MET and DOE under legislation of the Environment Protection Act 1972. Local background activities include both residential and industrial processes. In addition, traffic may influence the site as well as the Federal Highway is about 400 m away.

Overall, Peninsular Malaysia experiences relatively uniform temperature, high humidity and copious rainfall throughout the year. Wind flow distinguishes the seasons for Peninsular Malaysia, namely the South-West (SW) monsoon, the North-East (NE) monsoon and two shorter periods of inter-monsoons (INT.2 and INT.1) (METMalaysia, 2013). The SW monsoon is normally known as the dry season (middle May until middle September) in which haze is expected to occur while the NE monsoon is known as the wet season (early November until middle March) with the highest rainfall and the possibility of flooding. In this study, air pollution episodes are defined considering

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$PM_{2.5}$ mass ($> 40\,\mu g\,m^{-3}$) and the Air Pollution Index (API) (> 50), hereafter defined as HAZE samples. Local wind rose, seasonal regional synoptic wind field and biomass fire hotspots were given in Fig. S1 in the Supplement. The average temperature (T) at the site during the sampling campaign was $28.49 \pm 1.19\,^{\circ}C$ and the average relative humidity (RH) was $71.23 \pm 7.91\,\%$. Following the trend of T and API, wind speed was highest during the SW monsoon at an average of $1.39 \pm 0.19\,m\,s^{-1}$ and lowest during the NE monsoon at $1.2 \pm 0.17\,m\,s^{-1}$ with an annual average of $1.29 \pm 0.19\,m\,s^{-1}$. Rainfall was lowest during SW (6.27 ± 10.63 mm) and highest during NE (15.13 ± 22.69 mm). Overall, the main wind direction for the site was south-easterly, that is East-South-East (ESE), South-East (SE) and South-South-East (SSE). Details of the meteorological and gaseous pollutants for each season are given in Table S1 in the Supplement.

2.2 Aerosol sampling

The sampling was conducted from 4 August 2011 to 17 July 2012, for eight consecutive days every month (inclusive of one field blank) during a one-year sampling period. Sampling (24 ± 1 h; around 09:00 to 09:00) was performed using a high volume $PM_{2.5}$ sampler (Tisch Environmental, Inc.; Model TE-6070V-2.5-BL; USA) running at $1.13\,m^3\,min^{-1}$. Filter media used for sample collection were quartz micro-fibre filters (Whatman, QMA catalogue number 1851-865, UK) and were used directly without pre-cleaning. Before sampling, QMA filters were prepared such that every filter was wrapped with aluminium foil and pre-baked at $500\,^{\circ}C$ for 3 h inside a furnace (Nabertherm; Model L 5/11; Germany). In order to minimise the influence of water adsorption, loaded and unloaded QMA filters were equilibrated for 48 h in a desiccator and below 25 % RH prior to weighing. Aerosol masses ($PM_{2.5}$ mass) were deduced by weighing filter papers before and after sampling using a 5-Digit microbalance (A&D; Model GR-202; USA) with 0.01 mg sensitivity. A total of 94 filters (extra one sampling day for June 2012) were collected including 12 fields blank (one for each month). The samples were stored at $-18\,^{\circ}C$ in a freezer prior to analysis.

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2.3 Chemical analyses

2.3.1 Major ions

For the purpose of soluble ion analysis, one strip (2.54 cm × 20.32 cm) of loaded filter paper was used. The portion was cut into smaller pieces (1 cm × 1 cm) directly into a 50 mL conical flask. 20 mL of ultra-pure water, UPW (Hach, Millipore Direct-Q 3 UV System; USA) with a resistivity of 18.2 MΩ were added and the flask capped with a stopper. For sonication extraction purposes (60 °C; 60 m), an ultrasonic bath (Elma Schmidbauer GmbH; Elmasonic S40; Germany) was used. The solution was subsequently filtered through 0.2 µm 25 mm Acrodisc filters (Pall; Part number 4612; USA) using a 20 cc mL^{-1} Terumo syringe directly into a 25 mL volumetric flask, class A. UPW was added to the solution to the mark. The solutions were then directly transferred into two sets of 12 mL centrifuge tubes for separate anion and cation analysis. The extracted solutions were stored overnight in a refrigerator at 4 °C before analysis using ion chromatography (IC). The analysis took place within 48 h of extraction. Anion (F^-, Cl^-, NO_2^-, Br^-, NO_3^-, PO_4^{3-}, SO_4^{2-}) were analysed using a Metrohm 882 Compact IC plus 1 equipped with column type Metrosep A Supp 5 – 150/4.0 (Metrohm; USA) while a Metrohm 733 IC Separation Centre (Metrohm; USA) was used for cation analysis (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}). A six-point calibration (0.5, 1, 2, 5, 10 and 20 ppm) was used. The method detection limits (MDL) were calculated based on three times the standard deviation of field blank ($n = 6$) while 1 ppm standard of Single Cation/Anion Standards (Certipur® Reference Materials for Ion Chromatography, Merck Millipore, Merck KGaA, Darmstadt, Germany) was use for percentage of recoveries purpose. The percentage recovery for all elements was between 86 and 131 % (Table S2).

2.3.2 Trace elements

For trace elements, microwave-assisted extraction using acid digestion (4 : 1 of HNO_3 and H_2O_2) was performed using a Milestone Microwave Laboratory System (Gemini

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BV; MLS-1200 Mega; Netherlands). For the digestion process, one strip (2.54 cm × 20.32 cm) of loaded filter was used with the following setting of time (m) and power (W) was used: (1) 1, 250; (2) 1, 0; (3) 8, 250; (4) 4, 400 and (5) 5, 650. The solution was subsequently filtered through 0.2 µm 25 mm Acrodisc filters (Pall Gelmann) using a 50 cc mL^{-1} Terumo syringe directly into a 50 mL Teflon volumetric flask. This solution was then topped up with UPW to the mark before transfer into a 60 mL high density polyethylene (HDPE) bottle for storage. These stocks were kept in a refrigerator at 4 °C before analysis. Analysis of the elements was carried out using inductively coupled plasma mass spectrometry, ICPMS (Perkin-Elmer Instrument; Model Elan 9000; USA). Two sets of solutions were prepared for two modes of ICPMS analysis as follows: solution (1) 50 mL stock of concentrated solution for elements with lower weight; and solution (2) further diluted 1 : 4 (50 mL concentrated: UPW) for heavier weight elements. Four point calibration curves were performed for each mode of analysis as follows: mode (1) 10, 20, 30 and 50 ppb for Ag, As, Cd, Cr, Li, Be, Bi, Cs, Co, Cu, Ga, Mn, Ni, Rb, Se, Sr, U and V; and mode (2) 125, 250, 500 and 1000 ppb for Al, Ba, Fe, Pb and Zn. MDL was estimated as three times the standard deviation of field blank ($n = 6$) while 1 ppm Multi-Element Calibration Standard 3 (Perkin Elmer Pure Plus, Perkin-Elmer; USA) was use for validation purpose. Percentage recoveries are based on SRM1648a Urban PM (National Institute of Standards and Technology, MD, USA) and these varied between 29 and 101 % (Table S2).

2.3.3 Black carbon

Black carbon (BC) concentration was determined using Smokestain Reflectometer with calibration (Diffusion Systems Ltd.; Model EEL 43M; UK). Five points throughout the filters were taken where the average was then used as the final measured percentage of reflectance for mass calculation. Additional explanations pertaining to this instrument and the calculation involved have been discussed elsewhere (Wiwolwattanapun et al., 2011).

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2.3.4 Quality assurance

Details are provided in the Supplement.

2.4 Meteorological-gaseous measurements

All meteorological parameters and gaseous pollutants were obtained from the Air Quality Division of the DOE, Ministry of Natural Resources and Environment of Malaysia. The meteorological parameters included temperature (T), RH, wind speed (WS), wind direction (WD) and daily values of API readings while the gaseous pollutants were carbon monoxide (CO), ozone (O_3), sulphur dioxide (SO_2), nitrogen oxides (NO_x), nitrogen monoxide (NO) and nitrogen dioxide (NO_2). API for Malaysia is calculated based on five major air pollutants including SO_2, NO_2, CO, PM_{10} and O_3. These measurements were recorded at a station (registered station for the DOE Malaysia) less than 1 km south from our sampling location. Details of the monitoring equipment and procedures involved have been described by (Khan et al., 2015a). Daily rainfall readings, daily PM_{10} ($dp < 10\,\mu m$) and TSP (total suspended particulate) mass (high volume sampler) were obtained from MET of Petaling Jaya recorded at the sampling site.

2.5 Data analysis and modelling

2.5.1 Statistical and diagram plot

All descriptive and statistical analyses were carried out using either PASW Statistics for Windows, Version 18 (SPSS, 2009) or using Microsoft® Excel 2010 (Excel, 2010) with the statistical add-in XLSTAT Version 2014.3.04 (Addinsoft, 2014). Meteorological analysis for monsoonal effects was conducted with the application of several adapted analysis software packages. For wind vectors, the Grid Analysis and Display System (GrADS version 2.0.2) was used. The synoptic wind fields were plotted using a dataset (u, v – wind) downloaded from the National Center for Environmental Protection

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(NCEP)/National Center for Atmospheric Research (NCAR) (http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.pressure.html). The dataset downloaded was selected at 925 hPa (500 m) with a mapping covering latitude: −10°, 20° N, longitude: 90°, 120° E. For biomass hotspots, fire data from the Moderate-resolution Imaging Spectroradiometer (MODIS) representing the biomass burning hotspots in the specific area of interest was used. Data were downloaded from the National Aeronautics and Space Administration-Land Atmosphere Near Real-time capability for Earth Observing System (EOS)-Fire Information for Resource Management System (NASA LANCE FIRMS) fire archive (https://firms.modaps.eosdis.nasa.gov/download/request.php) in the range of 10° S to 20° N and 90° W to 120° E. These data were then appended on the map plotted using Igor Pro 6.22A (WaveMetrics, USA). In addition, 48 h backward trajectories were also included onto the same map using the Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT 4.9). To ensure consistency with the wind field, the trajectory release was chosen at about 925 hPa (500 m) with 6 h trajectory intervals were selected. For local wind roses (for each season), which were plotted using Igor Pro 6.22A (WaveMetrics, USA), data obtained from the DOE were used.

2.5.2 Chemical mass closure

Modified from Bressi et al. (2013), seven major groups were considered for the CMC calculations: sea salt (ss), dust, secondary inorganic aerosol (SIA), trace element (TE), BC, K^+ and also the unidentified portion of the filter mass. Due to our low Al element recovery (36 %) and lack of Si and S elements which are the dominant elements in soil from $PM_{2.5}$ (Rahman et al., 2011), the dust fraction is therefore calculated using a straightforward approach where nss-Ca^{2+} over the mineral contribution was used. The 11 % mineral dust mass contribution for the Klang Valley area calculated by the aforementioned study was employed for the calculation. Following the direct CMC nss-Ca^{2+} approach, we therefore exclude the major mineral dust elements (Al, Fe) to calculate the rest of trace element mass contribution.

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The overall calculations involved for the CMC were as follows:

$$[PM_{2.5}] = [\text{Sea salt}] + [\text{Dust}] + [\text{SIA}] + [\text{TE}] + [\text{BC}] + [K^+] + [\text{Unidentified}] \quad (1)$$

where,

$$[\text{Sea salt}] = [Na^+] + [Cl^-] + [Mg^{2+}] + [\text{ss-}K^+] + [\text{ss-}Ca^{2+}] + [\text{ss-}SO_4^{2-}];$$

with

$$[\text{ss-}K^+] = 0.036 \times [Na^+]; \quad [\text{ss-}Ca^{2+}] = 0.038 \times [Na^+];$$

and

$$[\text{ss-}SO_4^{2-}] = 0.252 \times [Na^+]$$

$$[\text{Dust}] = [\text{nss-}Ca^{2+}]/0.11$$

$$[\text{SIA}] = [\text{nss-}SO_4^{2-}] + [NO_3^-] + [NH_4^+];$$

with

$$[\text{nss-}SO_4^{2-}] = [SO_4^{2-}] - [\text{ss-}SO_4^{2-}];$$

"nss-" standing for "non-sea salt".

2.5.3 Enrichment factor

Due to the low recovery of Al, in this study we opted to use Fe as our reference element for the enrichment factor (EF) analysis. A study by Cesari et al. (2012) derived a two-threshold system of EF in which, for re-suspended soils, elements with an EF of smaller than two (2) were considered to be from crustal sources, EF of larger than four (4) were considered from an anthropogenic origin while those in between were considered of mixed origin. Following the recommendation of Cesari et al. (2012), we chose two as the cut-off point for the separation between anthropogenic and crustal origins.

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2.5.4 Source apportionment

A combination of PMF version 5.0 (PMF 5.0) and multi-linear regression (MLR) analysis was employed to determine source apportionment. Before use, all undetected data point were replaced by half of the MDL while data with value but below MDL was remain as it is. The results of the MLR were used to apportion the $PM_{2.5}$ chemical compositions in order to identify sources. Details of the PMF procedure used in this study are similar to our previous work as discussed in Khan et al. (2015b).

3 Results and discussion

3.1 $PM_{2.5}$ mass and its relations to meteorological and gaseous conditions

3.1.1 $PM_{2.5}$ mass variations

$PM_{2.5}$ measurement values are presented in Fig. 2 and Table 1. Overall, $PM_{2.5}$ mass ranged between 6 and 118 $\mu g\,m^{-3}$, with 43 % of the samples exceed the 25 $\mu g\,m^{-3}$ daily $PM_{2.5}$ guideline set by the WHO (WHO, 2006) and 21 % sample exceedance of the 35 $\mu g\,m^{-3}$ standard of 24 h $PM_{2.5}$ United States Environmental Protection Agency (US EPA) National Ambient Air Quality Standards (NAAQS) (USEPA, 2015). The highest daily value (118 $\mu g\,m^{-3}$) was measured during the SW monsoon, almost five times the WHO daily guideline and more than three times the 24 h US EPA NAAQS standards. This value was recorded during the haze episode in June 2012. Strong variability can be observed from the monthly and days averages of $PM_{2.5}$ mass (Fig. 2b, d, e). The month of June recorded the highest monthly average $PM_{2.5}$ mass (61 $\mu g\,m^{-3}$) followed by September (42 $\mu g\,m^{-3}$). Both months were during the SW monsoon. The lowest monthly average of $PM_{2.5}$ was in November with 17 $\mu g\,m^{-3}$ during the NE monsoon. Among the weekdays, Friday recorded the highest average value of $PM_{2.5}$ mass

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at 33 µg m^{-3} while lowest was on Wednesday (24 µg m^{-3}). Meanwhile, weekends on average recorded lower $PM_{2.5}$ mass (26 µg m^{-3}) compared to weekdays (29 µg m^{-3}).

$PM_{2.5}$ mass shows significant variability between the NE monsoon and the three other seasons (SW, INT.2 and INT.1). During the NE monsoon, only 17 % exceedance of the daily WHO guideline was recorded while for three other seasons, more than 50 % exceedance of the daily WHO guideline was recorded (Fig. 2c). The minimum exceedance of NE was due to high rainfall (precipitation) and low wind speed during this time. Juneng et al. (2009) and Rashid and Griffiths (1995) also reported similar observations of seasonal fluctuation of particulate concentration with minimal concentration during the rainy season of the NE monsoon. Most exceedance days occurred during the dry seasons of the SW monsoon and INT.2 (middle May until end of October) with 66 and 71 % exceedance, respectively. Similar observations of high exceedances during the SW monsoon dry season have been recorded for Peninsular Malaysia in general and the Klang Valley in particular (Rashid and Griffiths, 1995; Juneng et al., 2011; Norela et al., 2013; Tahir et al., 2013b; Amil et al., 2014). Higher mass concentrations during the dry season were also seen in other SEA (Kim Oanh et al., 2006; Lestari and Mauliadi, 2009) and Asia cities (Reid et al., 2013). It is important to note that haze events always occur during the SW monsoon (Fig. 2a), thus it is anticipated that they will directly affect the SW overall mass concentration ($PM_{2.5}$). However, the ANOVA analysis showed that HAZE is significantly different from the SW monsoon on an overall perspective ($p = 0.003$). This is perhaps due to short pollution episodes (HAZE) compared to the long period of the SW monsoon. HAZE events for this study averaged at 61 ± 24 µg m^{-3}, higher ompared to the 2011 haze episode documented for Bangi area at 48.32 ± 10.07 µg m^{-3} by Amil et al. (2014).

The annual $PM_{2.5}$ mass for this study averaged at 28 ± 18 µg m^{-3}. This is almost triple (2.8 fold) the 10 µg m^{-3} WHO $PM_{2.5}$ annual guideline, 2.33 fold higher than the US EPA NAAQS $PM_{2.5}$ annual standard of 12 µg m^{-3} and 1.12 fold higher than the European Union (EU) $PM_{2.5}$ annual standards set at 25 µg m^{-3} (European Commission, 2015). $PM_{2.5}$ mass average for this study was comparatively very low compared to other big

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cities of Asia i.e. in India and China (Balakrishnaiah et al., 2012; Huang et al., 2013; Pachauri et al., 2013; Zhang et al., 2013) but variable when compared to other parts of the world (Dongarrà et al., 2010; Yin et al., 2010; Bressi et al., 2013; Squizzato et al., 2013); Table 2. On a local scale, the average value of $PM_{2.5}$ mass for the site was slightly higher than previous measurements carried out here during 2004–2008 ($27 \pm 10\,\mu g\,m^{-3}$) (Rahman et al., 2011) but lower compared to measurements carried out during 1998–2000 ($33\,\mu g\,m^{-3}$) (Keywood et al., 2003). Furthermore, our result for Petaling Jaya is comparatively higher than other parts of Peninsular Malaysia (Tahir et al., 2013b; Ee-Ling et al., 2015).

The mean $PM_{2.5}$ / PM_{10} ratio for the site was 0.72 ± 0.18 and the ratio for $PM_{2.5}$ / TSP was 0.46 ± 0.13; Table 1. PM_{10} / TSP ratio was 0.63 ± 0.12. The $PM_{2.5}$ / PM_{10} ratio at this site is comparatively higher than other studies in Asia as reported by Hopke et al. (2008) where most of the sites studied showed ratios of lower than 0.5. From the aforementioned study, however, an urban site in China and suburban site in Lembang, Indonesia recorded similar $PM_{2.5}$ / PM_{10} ratio to our result of more than 0.7. Our $PM_{2.5}$ / PM_{10} ratio was also in agreement with other cities in Europe. For example, Gehrig and Buchmann (2003) reported that the overall $PM_{2.5}$ / PM_{10} ratio for Switzerland (covering urban and suburban and rural areas) was between 0.75 and 0.76 while Gomišček et al. (2004) reported 0.7 for Austrian urban sites. Urban and industrial sites in Italy also showed comparatively similar values to our ratio with 0.67 ± 0.10 and 0.67 ± 0.12, respectively (Contini et al., 2014). Despite having different characteristics, the SW and NE monsoons still came out with similar values to the annual ratio at 0.72 ± 0.1 and 0.71 ± 0.13, respectively. This shows that perhaps rainfall and WD (which distinguishes the season) do not greatly influence the fine particles. Both intermonsoon seasons recorded the opposite mass concentration trend. INT.2 (average mass of $29 \pm 12\,\mu g\,m^{-3}$) showed a higher mass concentration than INT.1 (average mass of $23 \pm 8\,\mu g\,m^{-3}$) but a lower $PM_{2.5}$ / PM_{10} ratio (0.62 ± 0.17) than INT.1 (0.85 ± 0.4). This ratio of INT.1 is the highest $PM_{2.5}$ / PM_{10} ratio among all seasons, even higher than during HAZE episodes. HAZE-episode-only ratios were 0.74 ± 0.07. Annually, PM_{10}

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accounts for about 63 % of the TSP but the seasonal variability of the ambient aerosol at the site was high. During the dry season (the SW monsoon), ambient air at the site had particles in the ratio of approximately 50/50 coarse (PM dp > 2.5µm) to fine particles (PM dp < 2.5µm) ($PM_{2.5}$ / TSP = 0.5 ± 0.08). During INT.2 and the NE monsoon (wet season), the air was filled with more coarse particles, resulting in $PM_{2.5}$ / TSP ratios of 0.44 ± 0.12 and 0.4 ± 0.09, respectively. INT.1 and HAZE episodes on other hand both had a $PM_{2.5}$ / TSP ratio of 0.54, implying the ambient air contained almost the same portion of fine and coarse particles. With these ratios, we can conclude that fine particles are very significant in the ambient air of the Petaling Jaya urban-industrial area in Klang Valley. Similar observation on the significance of the fine particle were also reported for SEA cities (Kim Oanh et al., 2006). In addition, a very significant correlation between $PM_{2.5}$ and PM_{10} ($r = 0.963$; $p < 0.0001$) revealed the possibility of similar sources for both particles as shown by a previous study of the site during 2004–2008 (Rahman et al., 2011). This theory of having the same source when $PM_{2.5}$ and PM_{10} mass correlated significantly has been suggested by Tahir et al. (2013b) during a study carried out at the coastal area of Kuala Terengganu with a value of 0.58 ± 0.06 for the $PM_{2.5}$ / PM_{10} ratio.

3.1.2 Relationship between $PM_{2.5}$ and meteorological-gaseous influence

The Pearson correlation revealed that $PM_{2.5}$ mass on an annual basis was significantly influenced by meteorological and gaseous parameters; Table 3. Among the parameters, API strongly correlated with $PM_{2.5}$ mass ($r = 0.763$; $p < 0.001$). Since the Malaysian API includes PM_{10}, this result was anticipated due to the high ratio of $PM_{2.5}$ / PM_{10} (0.72). The $PM_{2.5}$ mass was positively correlated with T ($r = 0.31$; $p = 0.005$) and negatively correlated with RH ($r = -0.314$; $p < 0.005$). Having used wind flow to distinguish the season for Malaysia, the WS influence towards the $PM_{2.5}$ mass was as expected ($r = 0.274$; $p < 0.05$). However, rainfall and WD did not significantly correlate with $PM_{2.5}$ mass at the site. With an exception of NO_x, all other gaseous parameters were found to significantly influence the $PM_{2.5}$ mass. CO and

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NO_2 were significantly positively correlated with $PM_{2.5}$ ($p < 0.0001$) at $r = 0.471$ and $r = 0.473$ respectively, indicating a combustion-related traffic source. The significant positive correlation between $PM_{2.5}$ and SO_2 ($r = 0.324$; $p < 0.005$) further supports this. NO was the only gaseous parameter that had a negative relationship with $PM_{2.5}$ mass ($r = -0.262$; $p < 0.0001$). O_3 on the other hand showed a significant positive correlation with $PM_{2.5}$ mass at $r = 0.298$ ($p < 0.01$).

On a seasonal scale, daily $PM_{2.5}$ mass during all seasons appeared to be affected by the gaseous parameters but not meteorological conditions. $PM_{2.5}$ mass during the SW monsoon, which also known as the dry season, was strongly correlated with CO ($r = 0.687$; $p < 0.001$), O_3 ($r = 0.535$; $p < 0.005$), NO_2 ($r = 0.528$; $p < 0.05$) and API ($r = 0.748$; $p < 0.001$). NE (the wet season) showed strong correlations with SO_2 and NO_2 with $r = 0.654$ ($p < 0.001$) and $r = 0.711$ ($p < 0.001$), respectively. NO showed the least effect towards $PM_{2.5}$ mass. Both INT.2 and INT.1 correlated strongly with NO_2, $r = 0.851$ ($p < 0.001$) and $r = 0.874$ ($p < 0.001$), respectively. In addition, INT.2 also showed a significant correlation with NO_x ($r = 0.800$; $p < 0.001$) while INT.1 correlated strongly with CO ($r = 0.654$; $p < 0.05$) and API ($r = 0.705$; $p < 0.05$). HAZE episodes, as expected, were significantly correlated with CO ($r = 0.749$; $p < 0.05$), which is one of the key pollution tracers. With Malaysia having relatively uniform temperature, high humidity and copious rainfall throughout the year, minimal influence of meteorological parameters towards seasonal $PM_{2.5}$ mass variation is predicted. Rainfall showed no significant correlation with $PM_{2.5}$ mass even during the two seasons of the SW monsoon (dry season with low RH and rainfall, high WS) and the NE monsoon (wet season with high RH and rainfall, low WS). However, INT.2 showed a strong negative correlation with rainfall ($r = -0.733$, $p > 0.05$). This may be due to the transition period of the WD in between the two monsoons. For the $PM_{2.5}$–T relationship, all four seasons of Peninsular Malaysia shows positive correlations. HAZE events revealed a slight negative correlation between $PM_{2.5}$ mass and T. RH and $PM_{2.5}$ mass on the other hand, revealed negative relationships with three seasons (except INT.1) having low correlations. INT.1 showed the reverse relationship. However, HAZE events which

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occur during the SW monsoon, agree with the generic pattern of the SW monsoon $PM_{2.5}$-RH relationship. WS and WD on a seasonal scale showed no significant correlation towards $PM_{2.5}$ in all four seasons, even during the HAZE events. As mentioned earlier, the $PM_{2.5}$ / PM_{10} ratio for both major seasons (SW and NE) were almost the same at ~ 0.7 (Table 1). The $PM_{2.5}$ / TSP and PM_{10} / TSP ratios were different, however. During the SW monsoon ratios of 0.5 and 0.7 were observed, while during the NE monsoon ratios of 0.4 and ~ 0.6 were recorded for $PM_{2.5}$ / TSP and PM_{10} / TSP respectively. These ratios support the findings of meteorological parameters (rainfall, WS and WD) not significantly correlating with $PM_{2.5}$ mass variability with changing season at the site. Instead, results reveal that perhaps meteorological parameters only greatly influence the coarse particles (PM dp > 2.5 µm) but not fine particles at the site.

3.2 Chemical composition

Referring to Fig. 3a and Table S2, chemical compositions of $PM_{2.5}$ determined were water soluble ions (anions and cations), trace elements (including heavy metals) and BC for a total of 36 % of $PM_{2.5}$ mass. BC accounted for about 15 % (4.15 µg m^{-3}) of the $PM_{2.5}$ mass. The total anion mass measured was 1.67 µg m^{-3} (5.98 % of $PM_{2.5}$ mass) while the total cation mass was 1.75 µg m^{-3} (6.26 % of $PM_{2.5}$ mass) with a total cation to total anion ratio of 0.46 (Fig. S2). The trend for anions was: SO_4^{2-} > NO_3^- > PO_4^{3-} > Cl^- > Br^- > NO_2^- > F^- while the cation trend was: NH_4^+ > Na^+ > K^+ > Ca^{2+} > Mg^{2+}. The overall water soluble trend for this urban-industrial site was: SO_4^{2-} (39 % of water soluble ion; 23 % of IM mass) > NH_4^+ (29 % of water soluble ion; 17 % of IM mass) > Na^+ (9 % of water soluble ion, 5 % of IM mass) > K^+ (7 % of water soluble ion; 4 % of IM mass) > NO_3^- (6 % of water soluble ion; 4 % of IM mass) > Ca^{2+} > PO_4^{3-} > Mg^{2+} > Cl^- > Br^- > NO_2^- > F^-. Trace elements on the other hand accounted for about 8.6 % of $PM_{2.5}$ mass (2.41 µg m^{-3}) with the major elements Al (44 % of TE), Fe (42 %), Zn (8 %), and Pb (4 %). The rest of the trace elements were in the decreasing order of: Ba > Cr > Cu > Rb > Mn > V > Ni > As > Sr > Ag > Cd > Se > Ga >

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Cs > Bi > Co > Li > U > Be. It is notable that results for Pb, As, Cd and Ni in this study did not exceed any EU standard on air pollutants. The 8.6 % mass percentage of trace elements determined in this Petaling Jaya urban-industrial site is lower than the 14.07 % trace element recorded at Kuala Lumpur city (Rahman et al., 2011) but higher compared to Kuala Terengganu (Tahir et al., 2013b).

Chemical mass closure

For a better understanding of the $PM_{2.5}$ chemical variability on a seasonal scale, we constructed a CMC on proportions of all identified components; as illustrated in Fig. 3b. In general, the inorganic seasonal variability in $PM_{2.5}$ composition is relatively small with both primary and secondary components of $PM_{2.5}$ equally important. In this study, IM accounted for 19 % of $PM_{2.5}$ mass while BC accounted for 15 % for a total of the 34 % chemical composition determined. Therefore, 66 % was left unidentified which was presumed to be sulfur compounds (S) and organic matter. The components for the aforementioned inorganic portion were as follows: SIA (2.49 $\mu g\,m^{-3}$; 9 %) > dust (2.09 $\mu g\,m^{-3}$; 7 %) > TE (0.34 $\mu g\,m^{-3}$; 1 %) > sea salt (0.27 $\mu g\,m^{-3}$; 1 %) > K^+ (0.25 $\mu g\,m^{-3}$; 1 %).

SIA, a combination of nss-sulfate (nss-SO_4^{2-}), ammonium (NH_4^+) and nitrate (NO_3^-), in $PM_{2.5}$ maintained a similar portion throughout the year – between 8 to 10 %, with the highest portion during INT.2 and lowest during the HAZE. On an annual as well as a seasonal scale (including HAZE), nss-SO_4^{2-} (annual average = 1.29 $\mu g\,m^{-3}$; 5 % of $PM_{2.5}$ mass; 23 % of IM mass) was the major SIA component followed by NH_4^+ (annual average = 0.99 $\mu g\,m^{-3}$; 4 % of $PM_{2.5}$ mass; 17 % of IM mass) and NO_3^- (1.29 $\mu g\,m^{-3}$; 1 % of $PM_{2.5}$ mass; 4 % of IM mass). Total SIA on this site was 73 % of the total water soluble ions measured, which is lower compared to 79 % in Greece (Remoundaki et al., 2013) and 85 % in Italy (Squizzato et al., 2013). The value of nss-SO_4^{2-} (97 % of SO_4^{2-}) and nss-K^+ (96 % of K^+) in this study are almost the same as results from 2004–2008 by Keywood et al. (2003) at 98 % for both nss-SO_4^{2-} and nss-K^+ which is why

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SO_4^{2-} and K^+ were used for PMF SA instead of nss-SO_4^{2-} and nss-K^+. These results, however, are different from another local study (Tahir et al., 2013a) where nss-SO_4^{2-} and nss-K^+ at a coastal area only made up about 53 and 13 % respectively. Hence, we could draw a conclusion that the SIA at the site is influenced by anthropogenic activities rather than marine sources even though the Malacca Straits are only about 33 km away. Following the SIA trend, nss-SO_4^{2-} was highest (6 %) during INT.2 which is the start of the rainy season. Surprisingly, the SW and NE monsoons came out with the same nss-SO_4^{2-} portion in $PM_{2.5}$ (5 %) even though the two have significant differences in terms of meteorological conditions, especially WD and rainfall; refer to Fig. S1a, c for synoptic wind direction. NH_4^+ and NO_3^- also do not vary largely with season, portioned at 4 and 1 %; respectively. HAZE recorded the lowest NO_3^- portion in $PM_{2.5}$ at below half a percent while NH_4^+ was lowest during the NE monsoon. Also known as the acidity ratio, the neutralisation ratio (NR) was calculated to further investigate the acidity of the atmospheric aerosols; Table S1. The NR was calculated based on the ratio of the NH_4^+ concentration to the sum of the concentration of nss-SO_4^{2-}. The overall NR obtained for this site was 0.65 ± 0.31, indicating that more than half of the acidity of the aerosols was neutralised by the ammonium. This result is similar to an observation on the east coast of Peninsular Malaysia reported by Tahir et al. (2013a) where a substantial fraction of SO_4^{2-} was neutralised by NH_4^+. However, the result is low compared to the neighbouring country of Singapore with an NR of 0.96 (Balasubramanian et al., 2003). The NR ratio varied with season. The highest recorded NR was during the HAZE episodes with 0.85 ± 0.22. The rest of the values showed the following trend: INT1.1 (0.83 ± 0.76) > SW (0.72 ± 0.19) > NE (0.56 ± 0.11) > INT.2 (0.55 ± 0.07).

Trace elements, which are good indicators for anthropogenic factors, had a mass contribution of $0.34\ \mu g\,m^{-3}$ (1 %) on an annual basis with the following seasonal trend: INT.2 (2 %) > NE (2 %) > INT.1 (1 %) > SW (1 %) > HAZE (1 %). Referring to the EF analysis (Fig. S3), most of the metals studied can be assumed to originate from anthropogenic sources, i.e. Pb, Se, Zn, Cd, As, Bi, Ba, Cu, Rb, V and Ni. Other metals,

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i.e. Sr, Mn, Co, and Li, are considered to originate from crustal sources. Pb, Zn, Cu, Cd, V and Ni are reflecting the traffic sources. Co, Sr and Li are typical soil constituents (Pey et al., 2009). Following Kuo et al. (2007), the elements can be categorised based on the degree of enrichment which in this study the annual EF gives the following results: (1) highly enriched (EF ≥ 1000): Pb; (2) moderately enriched (100 < EF < 1000): Se, Zn and Cd; (3) slightly enriched (10 < EF < 100): As, Bi and Ba; and (4) minimally enriched (EF < 10): Cu, V, Ni, Sr, Mn, Co and Li. However, the seasonal results revealed a slight difference in a few elements (Rb, V and Ni); Fig. S3. It is noted that except during the SW monsoon, V and Ni seemed to not fit the anthropogenic source. Both elements are well known as heavy oil combustion indicators (Jiang et al., 2014). However, a study in Taiwan also argued that the $PM_{2.5}$ Ni element (EF value of 20.49) could be drawn from either soil or crustal sources while the $PM_{2.5}$ V element (EF = 6.8) was derived from the soil (Balakrishnaiah et al., 2012).

Dust as one of the minor mass components of $PM_{2.5}$ and averaged at 7 % on an annual basis. This component showed the highest percentage during INT.2 (9 %), decreased a little in the following NE monsoon (7 %), continued to decrease in the INT.1 (6 %) and increased back again during the following SW monsoon (9 %). The HAZE episodes, however, recorded the lowest dust portion in $PM_{2.5}$ at 6 %. The seasonal patterns of dust portions relate to the meteorological conditions. During the NE monsoon the wind is blown from the Siberian High (Siberian Anticyclone) over South-East Asia i.e. Southern-China (Indo-China), Cambodia, Vietnam and the Philippines while during the SW monsoon, the wind flow is from Australia and neighbouring countries, i.e. Singapura and Indonesia (especially Sumatera and Jawa Island); Fig. S1a, c.

Sea salts form only ~ 1 % of $PM_{2.5}$ mass on an annual scale confirming the findings of a previous study by Keywood et al. (2003). Seasonally, the percentage remains below 1 % except during INT.1 where the sea salt portion is highest (4 %). However, the specific percentage value still shows the difference where the NE, and SW monsoons, INT.2 and HAZE portion at 0.99, 0.38, 0.28 and 0.18, respectively. The low percentage of sea salt in $PM_{2.5}$ is similar to the findings of a study by Tahir et al. (2013a) which

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observed that nss-ionic species accounted for 88 % of the total ions associated with $PM_{2.5}$. $PM_{2.5}$ at this site is expected to have a low marine contribution because marine aerosol is typically associated with coarse particles as seen by Tahir et al. (2013b) and Almeida et al. (2005). Khan et al. (2010) also reported similar observations where the four major marine elements, Na^+, Cl^-, Ca^{2+} and Mg^{2+}, were dominant in coarse particles ($PM_{2.5-10}$ and $PM_{>10}$). K^+, which is normally recognised as the biomass burning indicator, represented only 1 % of $PM_{2.5}$ mass ($0.25 \pm 0.14\,\mu g\,m^{-3}$ on annual scale) regardless of the season change including the HAZE episodes.

Black carbon averaged at $4.15 \pm 0.64\,\mu g\,m^{-3}$ (15 % of $PM_{2.5}$ mass). The highest proportion was seen during the rainy season of the NE monsoon (21 %) and lowest during the dry season of the SW monsoon (11 %). The HAZE events showed a result of 8 %. The two inter-monsoons seasons recorded average values between the two major seasons. Also known as elemental carbon (EC) (Lanz et al., 2010), the BC result measured here is within the range of Malaysia's initial results on BC measured at the same site by Abas and Simoneit (1996). They found $9\,\mu g\,m^{-3}$ EC with $74\,\mu g\,m^{-3}$ of organic carbon (OC) in TSP samples (TSP mass of $300\,\mu g\,m^{-3}$) during haze episodes; while during normal days they found 8 and $14\,\mu g\,m^{-3}$ for EC and OC respectively from $74\,\mu g\,m^{-3}$ of TSP mass. The BC value for this study (annual = 15 %, HAZE = 8 %) was low compared to measurements at the same site during a 1998–2000 study by Keywood et al. (2003). However, our results showed a similar pattern where BC during HAZE events was lower by at least one third (1/3) compared to normal days (normal = 30 %, haze = 20 %). The BC portion here was also similar to measurements carried out in 2004–2008 by Rahman et al. (2011) at 15.8 %. On a regional scale, our results here are comparatively low compared to most other SEA cities as reported by (Reid et al., 2013).

On an annual scale, the unidentified components reached 66 % of the total $PM_{2.5}$ mass. Seasonal variability was observed, with the smallest in the NE monsoon (58 %) during the intensified rainfall with low WS while the largest portions were during HAZE (77 %), when rainfall was low with high WS. One reason for such high uncertainties in

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the CMCs is the lack of OC composition which is one of the major components in $PM_{2.5}$. Previous studies by Tahir et al. (2013b) and Cohen et al. (2004) also reported similar large unidentified portions of $PM_{2.5}$ which were presumed to be of organic composition. A large amount of OC (58 %) in $PM_{2.5}$ was also reported in India at Ahmedabad (Rengarajan et al., 2011) and in an urban-industrial area of Agra (Pachauri et al., 2013). Cheng et al. (2010) reported a very high carbonaceous portion of $PM_{2.5}$ in Hong Kong of ∼ 70 % for three roadside monitoring sites and ∼ 48 % at the ambient site. The portion of our IM and BC were also comparatively low compared to the previous study of the site by Keywood et al. (2003) with 28 and 30 % (normal days), respectively. A study by Remoundaki et al. (2013) revealed that sulfates and carbonaceous material are major fractions of $PM_{2.5}$, with 35 and 30 %, respectively. Considering only the identified composition, water absorption of water-soluble components may lead to a positive bias during weighing, even in a controlled environment (i.e. RH) (Speer et al., 1997). In addition, Zhang et al. (2013) mentioned that the volatilisation of NH_4NO_3 and organic matter may result in negative biases in the specific components. This is likely to happen during the major seasons of the NE and SW monsoons.

3.3 Source apportionment and its relation to meteorological-gaseous conditions

From our chemical composition dataset (80 samples × 31 elements), the PMF 5.0 model resolved five factors, identified as: (1) combustion of engine oil; (2) mineral dust; (3) mixed SIA and biomass burning; (4) mixed traffic and industrial; and (5) sea salt; Table 4 and Fig. 4a. The source contribution by each factor was summed up to estimate the predicted mass of $PM_{2.5}$. A strong and significant correlation ($R^2 = 0.901$) was observed as shown by a scatter plot, representing a regression of the predicted and measured $PM_{2.5}$ for SA analysis; Fig. 4b. Table 4 summarises the SA results of the relative contributions from each identified source to the $PM_{2.5}$ on a seasonal and annual basis. The dominance of each identified source largely varies with changing seasons, which is roughly consistent with the CMC, EF and stoichiometric analysis for

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a number of factors. Each of the factors is characterised by a chemical "fingerprint" which is a unique pattern of chemical species and their concentrations. In addition, we also describe the interpretation SA identified in time series analysis and its relation to meteorological and gaseous factors (Fig. 5).

3.3.1 Factor 1: combustion of engine oil (V, Sr, Ni, SO_4^{2-}, Ga, NH_4^+)

With an annual V / Ni ratio of 1.91, both elements indicate a major contribution of fuel oil combustion, identified in this study as factor 1. Vanadium in this factor accounts for 53 % of total V mass while Ni represents 51 % (of total Ni mass). Many studies have used both elements as combustion fuel oil indicators (Kowalczyk et al., 1982; Harrison et al., 1996; Ho et al., 2006; Pey et al., 2009; Jiang et al., 2014). Mueller et al. (2011) indicated that V and Ni were promising markers for ship engine exhaust while Gibson et al. (2013) identified a shipping emissions factor based on V, Ni and SO_4^{2-} following a study by Zhao et al. (2013). Since Port Klang (one of the major ports in Malaysia) is about 33 km from our sampling site, there is a possibility of ship emissions to contributing to this factor. However, a number of studies have recognised a combination of V, Ni and SO_4^{2-} in $PM_{2.5}$ as oil combustion or industry as their interpretation of the source (Viana et al., 2008), dependent on the area surrounding the site. With an average contribution of 17 % on an annual basis, this factor does not change significantly over the seasons. The SW, NE and INT.1 monsoons have roughly the same percentage of around 16–17 %. INT.2 however scores the highest at 24 % (V / Ni ratio = 2.36), triple the HAZE events at only 7 % (V / Ni ratio = 1.74). The slight inconsistencies of the percentage portion seasonally may be due to different batches of heavy oil and origins of crude oil, as discussed by Jiang et al. (2014) based on studies by Mueller et al. (2011) and Zaki et al. (1989).

Factor 1 seems to not be particularly affected by gaseous parameters or meteorological conditions; Table S3. Overall, API and this factor did not correlate well, with an exception during NE (r = 0.366; p = 0.047). WD is the only meteorological parameter that is significantly correlated with this factor, and this occured during SW (r = 0.581;

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$p = 0.007$) which may have resulted from HAZE ($r = 0.677$; $p = 0.045$). For gaseous parameters, factor 1 seemed to be influenced by gaseous parameters mostly during the NE monsoon, with significant positive correlations with CO ($r = 0.498$; $p = 0.005$), SO_2 ($r = 0.436$; $p = 0.016$), NO_x ($r = 0.471$; $p = 0.009$) and NO_2 ($r = 0.529$; $p = 0.003$). O_3 is the only gas that appears to have more than one season correlating significantly with this factor. A negative correlation was shown between this factor and O_3 during SW ($r = -0.605$; $p = 0.001$), while a positive correlation ($r = 0.796$; $p = 0.032$) was seen during INT.2. Annually, only O_3 and SO_2 have significant correlations with this factor at $r = -0.287$ ($p = 0.014$) and $r = 0.380$ ($p = 0.001$), respectively.

3.3.2 Factor 2: mineral dust (Al, Li, U, Fe, Co, Ca^{2+}, Sr, Mn, Mg^{2+})

Factor 2 makes up 14 % of the $PM_{2.5}$ mass (annual average). This factor was identified based on elements Al (77 % of the Al mass), Li (61 % of the Li mass), U (45 % of the U mass), Fe (40 % of the Fe mass), Co (38 % of the Co mass), Ca^{2+} (33 % of Ca^{2+} mass) and Mg^{2+} (28 % of Mg^{2+} mass), as shown in Fig. 4a. Researchers cite these elements as markers for a mineral dust source. For example, Al and Fe were cited by Viana et al. (2008), Li and Fe by Pey et al. (2009) while Al and Fe by Balakrishnaiah et al. (2012). Mustaffa et al. (2014) reported a mineral dust source based on the presence of Ca^{2+} while Zhang et al. (2011) have used Mg^{2+} and Ca^{2+} as the indicators for a mineral dust factor. Ca^{2+} and Mg^{2+} were also used to classify crust ions in $PM_{2.5}$ (Wang et al., 2005). Fe also represents typical soil constituents and/or crustal combustion (Ho et al., 2006; Aldabe et al., 2011).

During three consecutive seasons of the year, i.e. the SW, INT.2 and NE monsoons (middle May 2011 until early March 2012), the mineral dust source portion remains about the same at around 15–16 % of the $PM_{2.5}$ mass. However, during the following inter-monsoon (INT.1), this factor was reduced to half at 7 %. The HAZE events on the other hand recorded the highest portion of this source with 19 % of the $PM_{2.5}$ mass. The reason is shown from the relationship between this factor and meteorological factors during this time period. HAZE seems to be affected by a few gaseous parameters i.e.

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NO_x and NO with $r = 0.650$ ($p = 0.042$) and $r = 0.698$ ($p = 0.025$), respectively. Annually, only SO_2 and NO_2 have significant relationships with factor 2, $r = 0.345$ ($p = 0.005$) and $r = 0.26$ ($p = 0.035$). Except during both inter-monsoons, mineral dust had a significant relationship towards T (strong positive correlation) and RH (strong negative correlation) including HAZE which happens during the SW monsoon. This may be the reason why the SW monsoon and factor 2 records the strongest correlation compared to other seasons in Malaysia, with $r = 0.673$ ($p < 0.001$) towards T and $r = -0.734$ ($p < 0.001$) towards RH.

3.3.3 Factor 3: mixed SIA and biomass burning (NH_4^+, Se, K^+, SO_4^{2-}, Rb)

The combined sum of ammonium sulfate and ammonium nitrate represents the secondary inorganic contribution to the $PM_{2.5}$ mass. This study is clearly dominated by ammonium sulfate. The potassium ion (K^+) on the other hand is an indication of major soil elements, usually from biomass burning. Echalar et al. (1995) has indicated that potassium (K) may be considered a good tracer for the flaming phase of forest fires. Watson and Chow (2001) reported that 85 % of the K is in the soluble form K^+, which is consistent with most vegetative burning profiles. Due to this established relationship, studies have used K in $PM_{2.5}$ as an indication for biomass burning (Song et al., 2006b; Santoso et al., 2008; Srimuruganandam and Shiva Nagendra, 2012b). However, a study by Pachon et al. (2013) suggested using total K in $PM_{2.5}$ in PMF can overestimate the contribution of biomass burning. Instead, they suggest using their newly-developed potassium associated with biomass burning (K_b) which proved to be a good indicator of biomass burning. Similarly, K^+ in PM was seen in many studies as a marker of biomass origin, either in the European region (Reisen et al., 2013) or SEA region (Tahir et al., 2013b; Wahid et al., 2013; Mustaffa et al., 2014; Ee-Ling et al., 2015). Reche et al. (2012) reported that K^+ from biomass burning was mostly emitted in the fine fraction of PM rather than coarse particles. Their biomass burning factor was indicated based on PMF results with 66 % of soluble K in addition to 84 % total variance of levoglucosan and 15 % OC. Characterised by high levels of NH_4^+ (59 % of NH_4^+

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mass), SO_4^{2-} (46 % of SO_4^{2-} mass) and K^+ (49 % of K^+ mass), the third and biggest factor for this site was identified as a mix of SIA and biomass burning and makes up 42 % of the $PM_{2.5}$ mass on annual basis. Studies by Mooibroek et al. (2011), Zhang et al. (2013), Almeida et al. (2005), Yin et al. (2010) and Song et al. (2006a) also identified a major contribution by the secondary aerosol fraction to $PM_{2.5}$.

In this study, highest mass contribution of factor 3 was observed during the SW monsoon (51 %) during which haze episodes normally occur. The rest of the year i.e. INT.2, NE and INT.1 represent 35 % or less of the $PM_{2.5}$ mass i.e. 35, 34 and 26 % respectively. This can be explained from PMF-CMC correlation matrix results where, contrary to other seasons, INT.1 during this time period only significantly correlated with SO_4^{2-} but not with NH_4^+ or K^+. The HAZE events in this study represented 63 % of the $PM_{2.5}$ mass. The time series (Fig. 6c) shows that this factor's elevated contribution occurred during a period from July until the end of October which is when the haze episodes normally occur. The HYSPLIT back trajectories analysis traced back the mass from the HAZE samples to Sumatera, i.e. Palembang during the 2011 episode and Palembang/Pekan Baru for 2012 episode; Fig. S4 a(ii), b(ii). This strongly suggested that during the period of the SW monsoon, the mass contribution of SIA and biomass factor could originate from long-range transport (regional influence) in addition to local agricultural and/or anthropogenic activities.

As shown by the factor-gaseous-meteorological correlation results, this factor on an annual scale seems to not correlate well with meteorological parameters, except for API and T. Season-wise, only API correlated well with this factor during SW and INT.2. However, on an annual scale, gaseous parameters showed varied relationships. CO, O_3, and NO_2 showed significant positive correlations towards this factor 3 while NO revealed a significant negative correlation. Season-wise, only the SW monsoon showed a strong relationship with this factor. The SW monsoon, with highest mass contribution of this factor, had significant ($p < 0.05$) positive correlations with CO, O_3, and NO_2 at $r = 0.612$, $r = 0.597$ and $r = 0.422$, respectively. The HAZE events on the other hand,

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although normally occurring during the SW monsoon, did not share these relationships. This factor during HAZE only correlated strongly with WS ($r = -0.678$; $p < 0.05$).

3.3.4 Factor 4: mixed traffic and industrial (NO_3^-, Pb, NO_2^-, Zn, As, Bi, Cd, BC, Se, Rb)

Dominated by NO_3^- (69 % of NO_3^- mass), Pb (58 % of Pb mass), NO_2^- (58 % of NO_2^- mass), Zn (55 % of Zn mass), As (51 % of As mass), Bi (47 % of Bi mass), Cd (44 % of Cd mass) and BC (38 % of BC mass), factor 4 was identified as mixed traffic and industrial sources with an average contribution of 10 % on an annual scale. As shown in Table 4 and illustrated in Fig. 5c, this factor varied with changing seasons. High contributions were seen from middle September until March during INT.2 (19 %) and NE (20 %) and very low contributions were seen during SW (4 %) and INT.1 (6 %) from April until September. HAZE appears to not to have significantly contributed to this factor with only 3 % mass contribution. Most of the trace elements in this factor are related to both traffic (Pb, Zn) and industrial emissions (As, Ni) (Fang et al., 2003; Querol et al., 2007). Pb and Zn are enriched in both vehicular emissions and also industrial emissions (Song et al., 2006a; Wåhlin et al., 2006; Querol et al., 2008; Pey et al., 2009; Thurston et al., 2011; Srimuruganandam and Shiva Nagendra, 2012b, a). EF results further suggest the Pb, Zn, As, Cd and Bi originated from anthropogenic sources. Malaysia has banned the use of Pb in petrol since 1996, indicating that the element is not originating from leaded petrol vehicle emissions. Thus, we exclude the influence of leaded petrol on this factor. Pastuszka et al. (2010) explain Pb mass as re-suspended road dust while Heal et al. (2005) explain Pb as road traffic emissions. Ewen et al. (2009) suggested that apart from the wear and tear of tyres, Cd is mainly emitted from the combustion of diesel fuel and oil or lubricants. Arsenic (As) mainly comes from industrial sources (Sánchez de la Campa et al., 2008; Stortini et al., 2009). Additionally, BC is an established tracer for primary anthropogenic emissions where its variability reflects changes in source strength, long-range transport and atmospheric mixing characteristics (Viidanoja et al., 2002). BC also is a major component of the $PM_{2.5}$ associated with road

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traffic emissions (Richmond-Bryant et al., 2009; Doumbia et al., 2012) and fuel oil combustion (Meyer, 2012; Zheng et al., 2012). Park et al. (2002) reported that the varying traffic and meteorological conditions of a site as well as the distance of the sampling equipment from the road traffic source will strongly influence the BC concentration. Data from the Malaysian Institute of Road Safety Research (MIROS) recorded a total of 342 279 vehicles in 24 h for the Federal Highway in October 2011 (Ministry of Works, 2011) which is near to our sampling station. During the peak hour of 08:00 to 09:00, 24 016 vehicles were recorded on this road. Previous studies have shown that road traffic can make substantial contributions to particulate mass concentrations in the Klang Valley area (Awang et al., 2000; Afroz et al., 2003; Rahman et al., 2011; Abdullah et al., 2012; Wahid et al., 2013; Ee-Ling et al., 2015; Khan et al., 2015b). NO_3^- and NO_2^- could also possibly come from the secondary aerosol of ammonium nitrate from anthropogenic activities in the surrounding area.

With NO_3^- and NO_2^- ions in the factor, a relationship between this factor and gaseous elements is anticipated. On an annual scale, NO, NO_x and NO_2 have shown significant positive correlations with this factor with $r = 0.428$ ($p < 0.001$), $r = 0.459$ ($p < 0.0001$), and $r = 0.311$ ($p = 0.008$) respectively, indicating a traffic emissions source. WS showed a significant negative relationship with this factor ($r = -0.39$; $p < 0.001$). Season-wise, following high mass contribution, INT.2 showed significant correlation with NO_x and NO_2 with $r = 0.774$ ($p < 0.05$) and $r = 0.766$ ($p < 0.05$), respectively. On the hand, NE as the dry season showed correlation with O_3 ($r = -0.351$; $p < 0.05$) and WS ($r = -0.507$; $p < 0.05$). Beckerman et al. (2008) reported that even though the level of NO_2 decay increases with increasing distance from the highway (at ~ 300 m), $PM_{2.5}$ still correlated strongly ($r > 0.7$; $p < 0.05$) with NO_2, NO and NO_x. They also found out that NO_2 still shows a strong association with $PM_{2.5}$ even with the potential of meteorological influences on the correlations. Pey et al. (2009) identified vehicle exhaust emissions based on high loadings of NO and CO in the principle components. A study in Korea by Park et al. (2002) concluded that BC is strongly correlated with CO and NO_x which can be further used as a vehicle emission tracer for the Seoul urban

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area. In addition, they also found that a $PM_{2.5}$-BC regression towards WS was negative, which is similar to our findings. These arguments further confirm the significance of our source type.

3.3.5 Factor 5: sea salt (Na^+, Cl^-, Mg^{2+}, Ca^{2+})

Making up an average of 17 % on an annual basis, sea salt was identified as factor 5 and was characterised by Na^+ (72 % of Na^+ mass), Cl^- (55 % of Cl^- mass), Mg^{2+} (45 % of Mg^{2+} mass) and Ca^{2+} (34 % of Ca^{2+} mass). Yin et al. (2005) identified sea salt based on primary marine aerosol of Na^+ and Cl^- in Ireland. Koçak et al. (2011) also used Na^+ and Cl^- to identify an aged sea salt factor for Istanbul. A study by Kim and Hopke (2008) defined a sea salt source by the high concentration of Na^+ and Cl in $PM_{2.5}$ while Begum et al. (2004) identified a sea salt factor based on Na and Cl elements in $PM_{2.5}$, measured by particle-induced x-ray emission. As shown in the time series (Table 4) and illustrated in Fig. 6c, the sea salt factor is seasonally high during INT.1 (45 %), April until early May. The other time periods were in the following mass contribution trend: NE (15 %) > SW (13 %) > HAZE (8 %) > INT.2 (6 %).

The understanding of the sea salt contribution during INT.1 requires some extended analysis. To investigate this, we carried out further stoichiometric analysis on the selected elements. The ratio of Mg^{2+} / Ca^{2+} on an annual scale was 0.11 while the seasonal ratios were: SW = 0.01, INT.2 = 0.08, NE = 0.07 and INT.1 = 0.24. The Cl^- / Na^+ ratios for all seasons were: SW = 0.11, INT.2 = 0.06 and NE = 0.14 and INT.1 = 0.04, with an overall annual ratio of 0.06. From these results, it is obvious that INT.2 contributed more Ca^{2+} and Na^+ with higher occurrences of chloride loss or the "chlorine deficiency" phenomenon compared to other seasons. According to Song and Carmichael (1999), chlorine in fine particles is almost exhausted in just 24 h. Khan et al. (2010) have reported that Cl loss in their study area is due to high humidity. Since Peninsular Malaysia is at the equator with very high T and RH, "chlorine deficiency" is a valid explanation. A similar observation of a low Cl^- / Na^+ ratio was also reported for Kuala Terengganu, Malaysia, at 0.02 (Tahir et al., 2013b). The $PM_{2.5}$ marine influence

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towards the sea salt factor has also been discussed elsewhere (Almeida et al., 2005, 2006).

The sea salt factor at this site seems to not have been influenced by meteorological conditions or the gaseous parameters. With the highest mass contribution of all seasons, sea salt during INT.1 showed a significant relationship ($p < 0.05$) with some gaseous parameters, i.e. CO, NO_x and NO_2 at $r = 0.694$, $r = 0.643$ and $r = 0.641$, respectively. T correlated with sea salt but only during the HAZE episodes ($r = 0.687$; $p < 0.05$) while rainfall showed a very strong relationship with sea salt during INT.2 with $r = -0.816$ ($p = 0.048$).

3.3.6 HAZE

As shown in Fig. 2a, two haze episodes occurred during our sampling period. The first episode occurred in September 2011 during the SW monsoon and the second episode occurred in June 2012, also during the SW monsoon. Since both episodes occurred during the same season, it is anticipated that both episodes have similar characteristics and therefore share the same origin. However our investigation revealed the two episodes to have quite different characters; Table S4. Although both episodes were most strongly influenced by the same source of mixed SIA and biomass burning, other sources did not follow the same trend. HAZE 2011 showed the following decreasing trend of sources: mixed SIA and biomass burning (81 %) > combustion of engine oil (10 %) > sea salt (5 %) > mineral dust (2 %) > mixed traffic and industrial (2 %) while HAZE 2012 showed the following: mixed SIA and biomass burning (56 %) > mineral dust (25 %) > sea salt (9 %) > combustion of engine oil (6 %) > mixed traffic and industrial (4 %). The PMF factor 3 of mixed SIA and biomass burning was further investigated through a correlation matrix between CMC and the source for a better understanding of the composition/characteristics; Table S5. HAZE 2012 showed a significant correlation between PMF factor 3 (mixed SIA and biomass burning) and CMC SIA with $r = 0.952$, $p < 0.001$. The PMF factor 3 during HAZE 2012 also showed significant correlations ($p < 0.001$) with SO_4^{2-} ($r = 0.963$), NH_4^+ ($r = 0.944$) and nss-

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SO_4^{2-} (0.965) but not with K^+. Further, the CMC SIA showed significant correlations with SO_4^{2-} ($r = 0.995$; $p < 0.0001$), NH_4^+ ($r = 0.997$; $p < 0.0001$) and K^+ ($r = 0.829$; $p = 0.011$). Therefore, we could conclude that PMF factor 3 (mixed SIA and biomass burning) during HAZE 2012 was in fact influenced by both SIA and biomass burning. HAZE 2011, however, indicated different sources. The PMF factor 3 did not have any significant correlation with CMC SIA, any of the CMC SIA elements or K^+. However, CMC SIA showed significant correlation with CMC SO_4^{2-} ($r = 1$; $p = 0.016$) and CMC NH_4^+ ($r = 1$; $p = 0.02$) but no significant correlation towards K^+. These results indicate that HAZE 2011 was mostly influenced by SIA alone and less so by biomass burning. With 10 % mass contribution from combustion of engine oil, HAZE 2011 could be concluded to have been influenced by anthropogenic activities including traffic. Besides SIA, a significant influence of mineral dust (25 %) and sea salt (9 %) showed that HAZE 2012 was greatly influenced by long-range transport. HYSPLIT backward trajectories for both HAZE episodes were traced back to Sumatera, Indonesia; Fig. S4 a(ii), b(ii). Further analysis showed that HAZE 2012 was more influenced by the meteorological and gaseous parameters compared to almost no significant correlation shown for HAZE 2011; Table S6. However, it is still not clear whether long-range transport did impact our HAZE episodes.

4 Conclusions

Our results revealed that fine particles are very significant in the ambient air of the Petaling Jaya urban-industrial area in the Klang Valley. The $PM_{2.5}$ mass averaged $28 \pm 18\,\mu g\,m^{-3}$ which is almost triple (2.8 fold) the WHO annual guideline. Our result is higher than reported for other parts of Peninsular Malaysia, but very low compared to other large Asian cities and variable when compared to other parts of the world. On a daily basis, the $PM_{2.5}$ mass ranged between 6 to $118\,\mu g\,m^{-3}$ with 43 % (samples) exceedance of the daily WHO guideline. Friday recorded the highest average value of

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$PM_{2.5}$ mass (33 µg m^{-3}) while lowest was on Wednesdays (24 µg m^{-3}). On average, weekends recorded lower $PM_{2.5}$ mass (26 µg m^{-3}) compared to weekdays (29 µg m^{-3}). The month of June during the dry season of the SW monsoon recorded the highest monthly average at 61 µg m^{-3} while November during the wet season of the NE monsoon recorded the lowest (17 µg m^{-3}). The NE monsoon is the only season that did not have more than 50 % exceedance of the daily WHO guideline.

In relation to meteorological-gaseous parameters, $PM_{2.5}$ mass on an annual scale showed the strongest relationship with API ($r = 0.763$; $p < 0.001$), explained by the $PM_{2.5}$ / PM_{10} ratio (0.72). As anticipated, $PM_{2.5}$ was positively correlated with T and WS but negatively correlated with RH. Rainfall and WD were not found to be significantly influential. With an exception of NO_x, all other gaseous parameters were found to significantly influence the $PM_{2.5}$ mass. CO, NO_2 and SO_2 were found to significantly correlate with $PM_{2.5}$ indicating a combustion-related traffic source. NO was the only gaseous parameter that had a negative relationship with $PM_{2.5}$ mass. O_3 at the site was also significantly correlated with $PM_{2.5}$ mass.

On a seasonal scale, daily $PM_{2.5}$ mass in all seasons was affected by the gaseous parameters but not meteorological conditions. The SW monsoon was found to have a significant relationship with CO, O_3, NO_2 and API while the NE monsoon was correlated with SO_2 and NO_2. Having relatively uniform T, RH and copious rainfall throughout the year, the small influence of meteorological parameters towards seasonal $PM_{2.5}$ mass variation was as anticipated. All four seasons showed positive correlations with $PM_{2.5}$ mass and T but the HAZE events revealed a slight negative correlation. The RH and $PM_{2.5}$ relationship was negative except during INT.1. Unexpectedly, rainfall, WS and WD did not significantly correlate with $PM_{2.5}$ mass variability with changing season even during the major seasons of the SW or NE monsoons. Further analysis on the $PM_{2.5}$ / PM_{10}, $PM_{2.5}$ / TSP and PM_{10} / TSP ratios revealed that meteorological parameters only greatly influence the coarse particles (PM > 2.5µm) but not so much on fine particles at this site.

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The $PM_{2.5}$ chemical compositions determined were anions, cations, TE and BC for a total of 36 % of the $PM_{2.5}$ mass. The total cation to total anion ratio was 0.46 with the ions in the decreasing trend: $SO_4^{2-} > NH_4^+ > Na^+ > K^+ > NO_3^- > Ca^{2+} > PO_4^{3-} > Mg^{2+} > Cl^- > Br^- > NO_2^- >$ F. TE analysis revealed Al, Fe, Zn, and Pb as the major elements. It is notable that results for Pb, As, Cd and Ni in this study did not exceed any EU standard on air pollutants. We further constructed CMC to better understand the seasonality variability in $PM_{2.5}$ composition. Our finding showed that both primary and secondary components of $PM_{2.5}$ are equally important, albeit with seasonal variability. The CMC components identified were: BC > SIA > Dust > TE > Sea salt > K^+. Seasonally, BC showed highest accountability during the NE monsoon and lowest during the SW monsoon but other CMC components did not vary largely with changing season. As for the SIA, the NR (0.65 ± 0.31) indicated that more than half of the acidity of aerosols was neutralised by ammonium. Further SIA components analysis revealed that SIA at the site was affected by anthropogenic activities rather than marine influences. The EF analysis further distinguished trace elements into two groups from anthropogenic sources (Pb, Se, Zn, Cd, As, Bi, Ba, Cu, Rb, V and Ni) and crustal sources (Sr, Mn, Co, and Li).

For SA purposes, we incorporated PMF 5.0 and MLR which revealed strong and significant correlations between the predicted and measured mass of $PM_{2.5}$ ($R^2 = 0.901$). Five factors were identified: (1) Combustion of engine oil; (2) Mineral dust; (3) Mixed SIA and biomass burning; (4) Mixed traffic and industrial; and (5) Sea salt; with an annual mean contribution of 17, 14, 42, 10 and 17 %, respectively. The dominance of each identified source largely varied with changing season but were roughly consistent with the CMC, EF and stoichiometric analysis for a few factors, accordingly. In addition to local anthropogenic activities, regional long-range transport was also influential. Further analysis on the HAZE episodes revealed different influences for the two different haze episodes. HAZE 2011 was mostly influenced by SIA but not so much by biomass burning, indicating more influence from anthropogenic activities (including traffic). Meanwhile, HAZE 2012 could be greatly influenced by long-range transport

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with large contributions from SIA, biomass burning, mineral dust and sea salt. HYSPLIT backward trajectories for both HAZE episodes traced the air masses back to Sumatera, Indonesia.

These results are connected to the urban-industrial background of the area, where gaseous parameters affect $PM_{2.5}$ mass both annually and seasonally. The results of our study clearly suggest that chemical constituents and sources of $PM_{2.5}$ were greatly influenced and characterised by meteorological and gaseous parameters which largely varied with season. However, our study is limited to inorganic and BC analysis, only a proportion of the overall mass. Further comprehensive assessment covering the organic portion and total elemental inorganic composition (i.e. total K, total Mg, total Na, total Ca, Si, S etc) is necessary for a complete composition dataset. In addition, it is suggested that particle number concentration (PNC) distribution should be incorporated into the chemical composition SA analysis as well. The potential source contribution function (PSDF) could also enhance the analysis of local and regional long-range transport. This would lead to improved analysis results and interpretation of the $PM_{2.5}$ dataset, which eventually will lead to better understanding of the fine particle variability here in Klang Valley.

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Table 1. Descriptive statistics of $PM_{2.5}$ mass and particulate matter (PM) ratio; unit: mean ± standard deviation (min–max). Remarks: SW = South-West monsoon; NE = North-East monsoon; INT.2 = Inter-monsoon 2; INT.1 = Inter-monsoon 1; HAZE = samples with $PM_{2.5}$ mass > 40 $\mu g\,m^{-3}$ and air pollution index (API) > 50.

Elements	ANNUAL 5 Aug 2011–18 Jul 2012 $n = 81$	SW 15 May–14 Sep $n = 29$	INT.2 15 Sep–30 Oct $n = 7$	NE 1 Nov–14 Mar $n = 35$	INT.1 15 Mar–14 May $n = 10$	HAZE $n = 11$
$PM_{2.5}$ ($\mu g\,m^{-3}$)	28 ± 17 (6–118)	38 ± 24 (14–118)	29 ± 12 (10–50)	21 ± 6 (6–35)	23 ± 8 (14–39)	61 ± 24 (40–118)
$PM_{2.5}$ / PM_{10}	0.72 ± 0.18	0.72 ± 0.10	0.62 ± 0.17	0.71 ± 0.13	0.85 ± 0.40	0.74 ± 0.07
$PM_{2.5}$ / TSP	0.46 ± 0.13	0.50 ± 0.08	0.44 ± 0.12	0.40 ± 0.09	0.54 ± 0.22	0.54 ± 0.07
PM_{10} / TSP	0.63 ± 0.12	0.7 ± 0.09	0.71 ± 0.06	0.57 ± 0.12	0.65 ± 0.09	0.73 ± 0.12

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Table 2. Comparison of $PM_{2.5}$ mass recorded in this study with other previous studies.

Location	$PM_{2.5}$ mass ($\mu g\,m^{-3}$)	Site description	Sampling period (24 h)	Reference
Petaling Jaya, Klang Valley, Malaysia	28 ± 17	Urban–industrial	5 Aug 2011–10 Jul 2012	This study
Kuala Lumpur, Klang Valley, Malaysia	30 ± 7 18 ± 3 10 ± 4	Urban Metropolitan Semi-urban Rural	Jan–Mar 2013	Ee-Ling et al. (2015)
Kuala Lumpur, Klang Valley, Malaysia	27 + 10	Urban	Jan 2004–Dec 2008	Rahman et al. (2011)
Kuala Terengganu, Malaysia	14 ± 7	Coastal, Sub-urban	Aug 2006–Dec 2007	Tahir et al. (2013b)
Petaling Jaya, Klang Valley, Malaysia	33	Urban–industrial	Dec 1998–Dec 2000	Keywood et al. (2003)
Gombak, Klang Valley, Malaysia	28	Urban–residential	Dec 1998–Dec 2000	
New Taipei City, Taiwan	22 ± 8	Urban Industrial	May 2011–Nov 2011	Gugamsetty et al. (2012)
Agra, India	140 ± 22 308 ± 52 91 ± 17	Urban Industrial Traffic Rural	Nov 2010–Feb 2011	Pachauri et al. (2013)
Paris, France	15 + 10 15 + 11	Urban Semi-urban	11 Sep 2009–10 Sep 2010	Bressi et al. (2013)
Qincheng, China	51 ± 18	Industrial complex	5–16 Aug 2009; 24 Jan–4 Feb 2010	Huang et al. (2013)
Beijing, China	135 ± 63	Urban	Apr 2009–Jan 2010	Zhang et al. (2013)
Venice, Italy	33 33 26	Urban Industrial Semi-urban	Mar 2009–Jan 2010	Squizzato et al. (2013)
Birmingham, UK	12 10	Urban Rural	May 2007–Apr 2008	Yin et al. (2010)
Palermo, Sicily, Italy	34 24	Metropolitan; Urban 1 Urban 2	Nov 2006–Feb 2008	Dongarrà et al. (2010)
Singapore	27 ± 10	Urban	Jan–Dec 2000	Karthikeyan and Balasubramanian (2006)

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Table 3. Pearson correlation matrix results between seasonal $PM_{2.5}$ mass and: **(a)** meteorological; and **(b)** gaseous parameters. Remarks: For meteorological parameters, API is air pollution index; T = temperature; RH = relative humidity; WS = wind speed; and WD = wind direction.

(a)	Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
	API	**0.763**[b]	**0.748**[b]	0.299	**0.473**[a]	**0.705**	0.531
	T	**0.310**	0.236	0.572	0.201	0.030	−0.050
	RH	**−0.314**[a]	−0.252	−0.495	−0.174	0.152	0.108
	WS	**0.274**	0.164	0.245	−0.030	0.192	−0.446
	WD	−0.131	−0.181	0.409	0.056	0.047	0.413
	Rainfall	−0.212	−0.246	−0.733	−0.052	−0.051	−0.178
(b)	Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
	CO	**0.471**[b]	**0.687**[b]	0.713	**0.488**[a]	**0.654**	**0.749**[a]
	O_3	**0.298**[a]	**0.535**[a]	0.427	**0.433**	0.378	0.449
	SO_2	**0.324**	0.141	−0.250	**0.654**[b]	0.627	0.445
	NO_x	0.058	0.112	**0.800**	**0.380**	0.588	0.192
	NO	**−0.262**	−0.309	0.701	0.086	−0.126	−0.285
	NO_2	**0.473**[b]	**0.528**[a]	**0.851**	**0.711**[b]	**0.874**[a]	0.599

Values in bold are different from 0 with a significance level alpha = 0.05;
[a] is when p values < 0.001 and
[b] p values < 0.0001.

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Table 4. Relative contribution of $PM_{2.5}$ sources from the positive matrix factorisation (PMF) analysis. Remarks: SIA = secondary inorganic aerosol.

Source contribution, $\mu g\,m^{-3}$ (%)	ANNUAL	SW	INT.2	NE	INT.1	HAZE
Factor 1: Combustion of engine oil	4.94 (17 %)	6.47 (17 %)	7.08 (24 %)	3.50 (16 %)	3.98 (16 %)	4.24 (7 %)
Factor 2: Mineral dust	3.95 (14 %)	5.49 (15 %)	4.58 (16 %)	3.18 (15 %)	1.62 (7 %)	11.28 (19 %)
Factor 3: Mixed SIA and biomass burning	11.72 (42 %)	19.05 (51 %)	9.99 (35 %)	7.44 (34 %)	6.21 (26 %)	36.92 (63 %)
Factor 4: Mixed traffic and industrial	2.93 (10 %)	1.30 (4 %)	5.42 (19 %)	4.28 (20 %)	1.29 (6 %)	1.85 (3 %)
Factor 5: Sea salt	4.67 (17 %)	4.98 (13 %)	1.80 (6 %)	3.20 (15 %)	10.76 (45 %)	4.62 (8 %)

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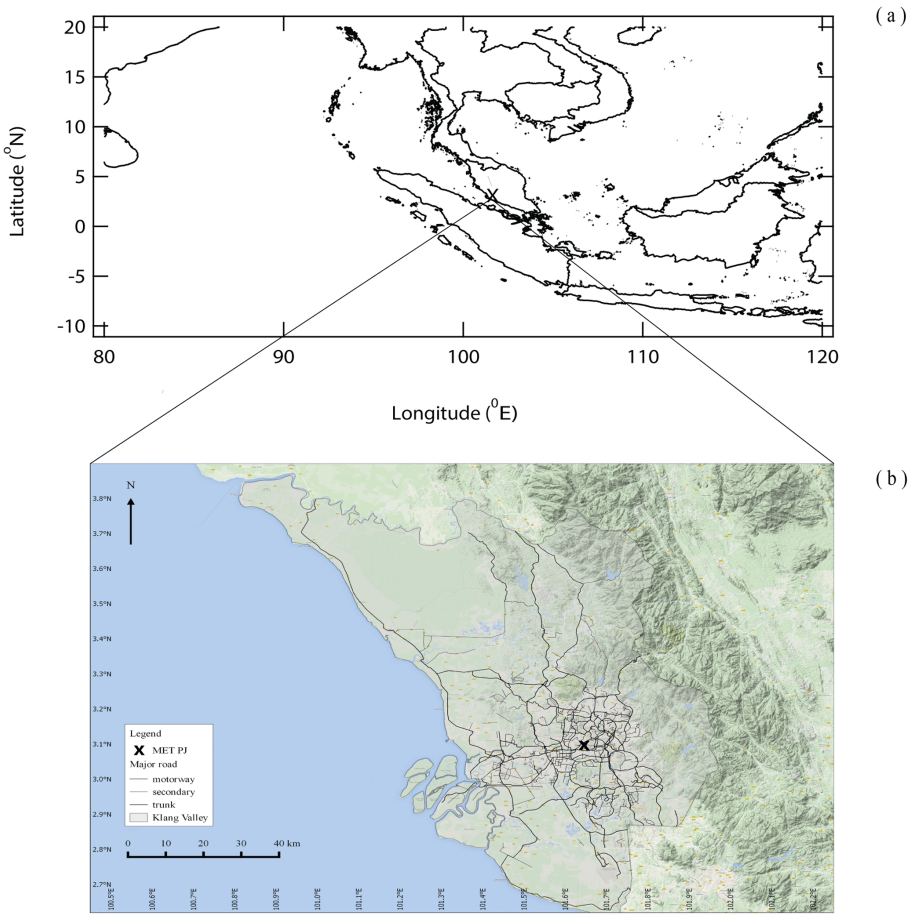

Figure 1. Location of the sampling site mark as "X" in: **(a)** the Southeast Asia region where the area is the boundaries of MODIS fire hotspot data used; and **(b)** the Klang Valley area in the Peninsular Malaysia.

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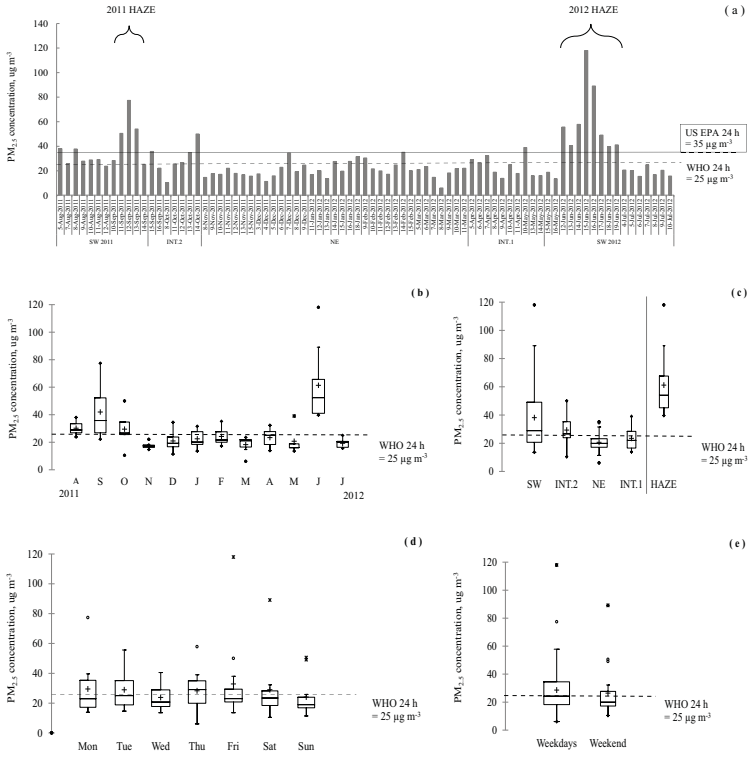

Figure 2. The $PM_{2.5}$ mass concentration on the: **(a)** daily basis; with box plot of the: **(b)** monthly; **(c)** seasonal; **(d)** days; and **(e)** weekdays/weekend. All figures were also subject to World Health Organisation (WHO) daily $PM_{2.5}$ guideline and United States Environmental Protection Agency (US EPA) daily $PM_{2.5}$ standard, accordingly.

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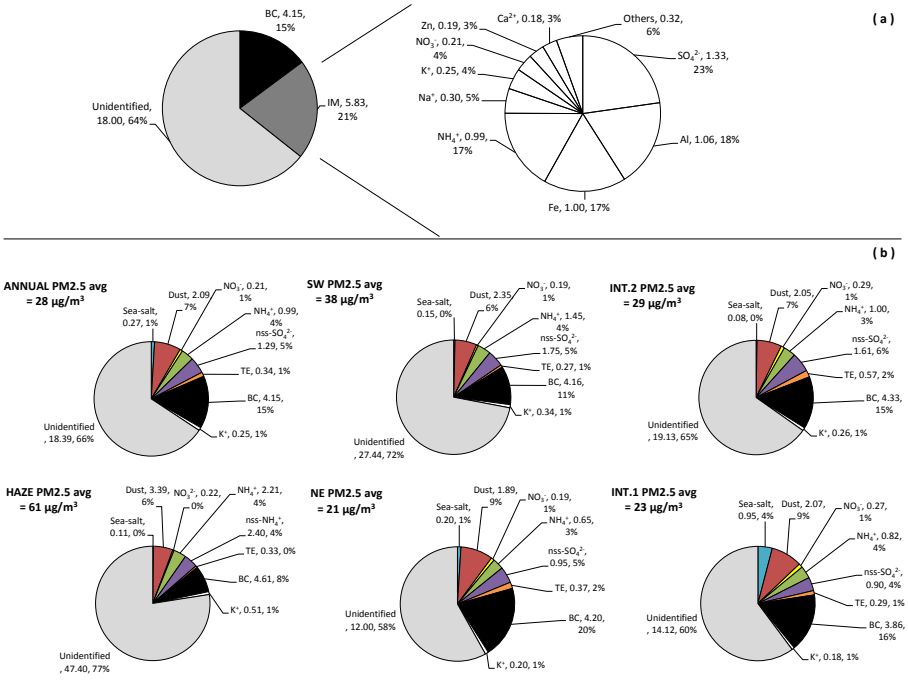

Figure 3. The composition of $PM_{2.5}$ displayed as [element; mass in $\mu g\,m^{-3}$; percentage in $PM_{2.5}$ mass] based on: **(a)** annual chemical composition determined where IM is the inorganic matter; and **(b)** seasonal chemical mass closure (CMC) components identified.

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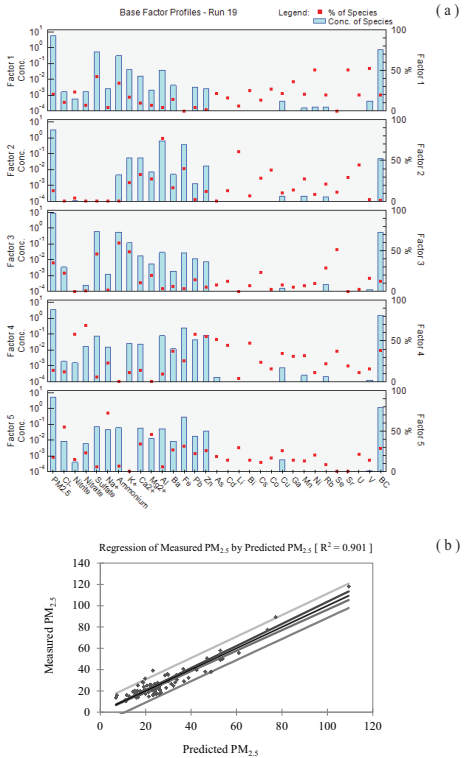

Figure 4. Source apportionment results from positive matrix factorisation (PMF) analysis: **(a)** source profile; and **(b)** regression plot between measured and predicted $PM_{2.5}$ mass. PMF factors were: factor 1 = combustion of engine oil; factor 2 = mineral dust; factor 3 = mixed secondary inorganic aerosol (SIA) and biomass burning; factor 4 = mixed traffic and industrial; and factor 5 = sea salt.

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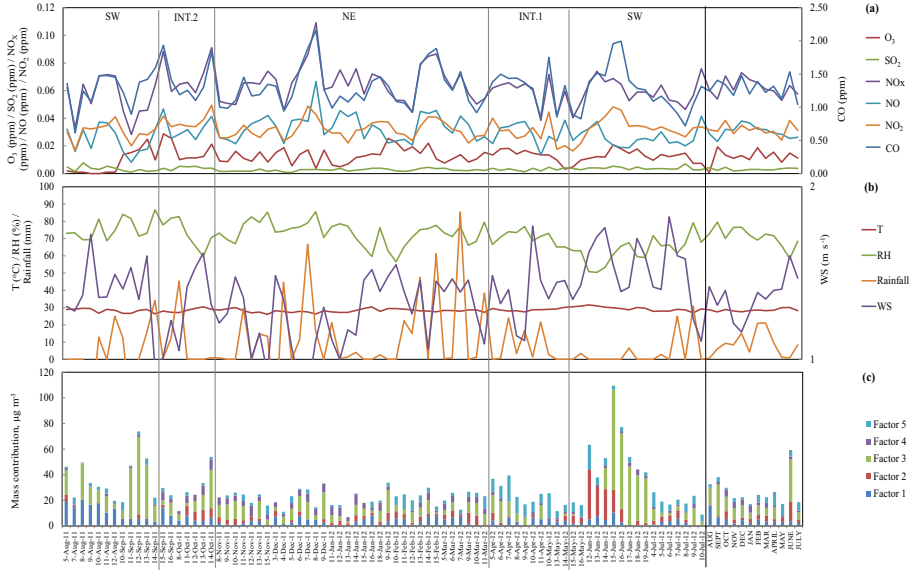

Figure 5. Time series of daily and monthly variations (left to right) of: **(a)** gaseous; **(b)** meteorological parameters; and **(c)** relative contribution of $PM_{2.5}$ sources.

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