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Seasonal variability of $PM_{2.5}$ composition and sources in the Klang Valley urban-industrial environment

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Abstract. This study investigates 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 1-year campaign from August 2011 to July 2012. This is the first paper on $PM_{2.5}$ mass, chemical composition and sources in the tropical environment of Southeast Asia, covering all four seasons (distinguished by the wind flow patterns) including haze events. The samples were analysed for various inorganic components and black carbon (BC). The chemical compositions were statistically analysed and the temporal aerosol pattern (seasonal) was characterised using descriptive analysis, correlation matrices, enrichment factor (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. In addition, secondary data of total suspended particulate (TSP) and coarse particulate matter (PM_{10}) sampled at the same location and time with this study (collected by Malaysian Meteorological Department) were used for PM ratio 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 the daily WHO guideline exceeded 43 % of the time. The north-east (NE) monsoon was the only season with less than 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 (particles with an aerodynamic diameter of greater than 2.5 µm) 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 in the decreasing order of (mass contribution) BC > secondary inorganic aerosols (SIA) > dust > trace elements > 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 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 con-

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tribution 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 vary greatly with season.

1 Introduction

Airborne particulate matter (PM) significantly impacts global climate (Jacobson, 2002; Vieno et al., 2014; Mallet et al., 2016), causing visibility degradation in both urban and less polluted 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). Fuzzi et al. (2015) revealed that climate–aerosol interaction, as well as effects of PM on human health and the environment, were underpinned by many new processes and development in the science. 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; Ross et al., 2013; Khan et al., 2016). The fine particles, which are composed of compounds of a range of volatilities, 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; Krewski et al., 2009; Tagaris et al., 2009; WHO, 2013).

The fraction and composition variability of fine particles ($PM_{2.5}$; particles with an aerodynamic diameter of less than 2.5 µm) are strongly influenced by seasonal meteorological factors, gaseous parameters and location. Megaritis et al. (2014) showed 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 changes and the background of an area (topography and local activities affecting anthropogenic and/or natural air pollution emissions) 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. For example, 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, WD and other meteorological parameters. Also, their chemical mass closure (CMC) components (i.e. soil dust, metallurgical industry, biomass burning and automobiles, sea salt and fuel oil combustion) at times were more significantly attributed to Indonesian forest fires compared to local traffic and industrial emissions. Ye et al. (2003) reported varied CMC elements (ammonium sulfate and nitrate, carbonaceous material, crustal components, potassium) 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 (dust, secondary sulfate, secondary nitrate, coal combustion, diesel and gasoline exhaust, secondary ammonium, biomass aerosol, cigarette smoke, vegetative detritus) showed distinct seasonal trends (Zheng et al., 2005). Indian $PM_{2.5}$ sources (i.e. motor vehicles, biomass burning, marine aerosol, tyre and brake wear, soil, secondary PM) 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. The study identified carbonaceous aerosol as the largest contributor, followed by ammonium sulfate, crustal material, sea salt and ammonium nitrate. 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. Receptor modelling 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. Receptor modelling uses temporal and chemical variations to separate total PM into different factors, where marker species are used to identify 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 the chemical mixture is positive matrix factorisation (PMF), first developed by Paatero and Tapper (1993). Subsequently, numerous other studies have employed this method in their $PM_{2.5}$ receptor modelling including many undertaken in the Asian region. For example, Begum et al. (2004) have successfully applied PMF on inorganic and BC data sets to lead to source identification for $PM_{2.5}$ in Bangladesh. Srimuruganandam and Shiva Nagendra (2012b) made an evaluation of $PM_{2.5}$ sources for Chennai city, India, using only inorganic (elemental) compositions. A study by Zhang et al. (2013) has successfully discussed the seasonal perspective of $PM_{2.5}$ sources (soil dust, coal combustion, biomass burning, traffic and waste incineration emissions, industrial pollution, secondary inorganic aerosol) in Beijing, China, using PMF on inorganic and organic data sets. Similar applications of PMF to apportion the sources of $PM_{2.5}$ have also been successfully carried out here in Southeast Asia (SEA). For example, Santoso et al. (2008) used inorganic and BC data sets to identify five major sources of $PM_{2.5}$ as biomass burning, soil, two stroke engine emissions, sea salt, secondary sulfate, motor vehicle emissions and road dust. A study by Rahman et al. (2011) also used similar chemical compositions for the SA analysis of $PM_{2.5}$ samples from the Klang Valley, which resulted in five sources: two stroke engine emissions, motor vehicle emissions, smoke/biomass burning, soil and industry. PMF was also effectively applied by Khan et al. (2015b) to their polycyclic aromatic hydrocarbon (PAH) data set to characterise the $PM_{2.5}$ for the semi-urban area of Bangi, Malaysia. The study revealed three main sources: gasoline combustion, diesel and heavy oil combustion, and natural gas and coal burning. 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). Due to limitations of a single model, applying more than one receptor model will enhance the SA analysis, leading to enhanced characterisation of an element and/or source and thus increasing the confidence in interpretations from the results. The study also reports that the most frequent combinations used for SA are principal component analysis (PCA) cluster analysis, PCA Lenschow, PCA chemical mass balance (CMB), PCA back-trajectory analysis, PMF UNMIX multilinear engine and CMB mass balance.

Reid et al. (2013) discussed in detail how the SEA region holds a complex relationship between geographic, socioeconomic, meteorological and aerosol microphysical factors. The review emphasised timing and location of sampling when trying to achieve a representation of the actual condition of the aerosol system, as the urban and industrial aerosol environments differ between urban centres. For example in Jakarta in Indonesia, two stroke engine vehicles, high emitters of particles and incomplete combustion products, were the major factor. Meanwhile, mobile sources are significant in Bangkok, Thailand, whereas Manila in the Philippines was significantly affected by diesel truck and bus emissions. Having said that, most urban centres in the region share the major sources of meat cooking and oil–gas–petrochemical industry activity as well as shipping influences. In addition, the region is also affected by haze episodes caused by biomass burning. Taking this into consideration, we conducted a 1-year assessment of $PM_{2.5}$ covering all four seasons (including haze events) to investigate its variability in the Klang Valley (urban-industrial) tropical environment. 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 (multi-linear regression) model in conjunction with the cluster analysis of back trajectory. All variables of $PM_{2.5}$ mass, their chemical compositions identified, as well as the sources predicted, were further analysed using correlation matrices with the meteorological–gaseous pollutants for comprehensive assessment.

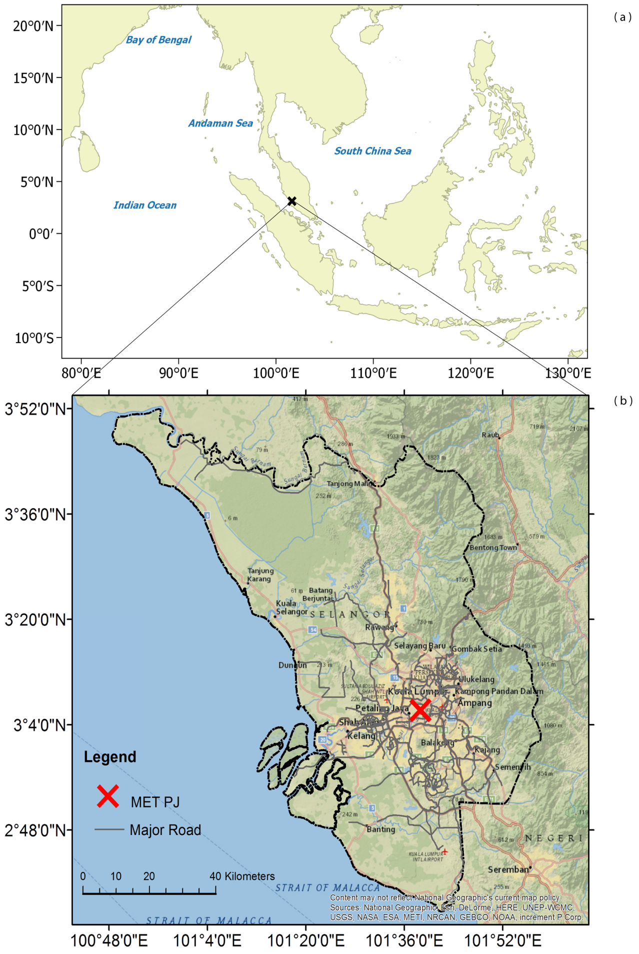

Figure 1. Location of the sampling site mark as "X" in **(a)** the Southeast Asian region and **(b)** the Klang Valley area in the Peninsular Malaysia.

2 Material and methods

2.1 Sampling site description

As shown in Fig. 1, 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. 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 stations representing the tropical region of the World Meteorological Organisation GAW network. This site is regarded as being representative of urban-industrial conditions, categorised according to criteria proposed by the Malaysia's MET and Department of Environment (DOE) under legislation of the Environment Protection Act of 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 ($\sim 28.5\,^{\circ}\mathrm{C}$), high humidity (more than 70 %) and copious rainfall (6.27–15.1 mm) throughout the year. Wind flow pattern distinguishes the seasons for Peninsular Malaysia, namely the south-west (SW) monsoon, the north-east (NE) monsoon and two shorter periods of intermonsoons (INT.2 and INT.1; METMalaysia, 2013). During the SW monsoon (usually established during the middle of May until the middle of September), the prevailing wind flow is generally south-westerly and light (below $7.72\,\mathrm{m\,s^{-1}}$). Known as the dry season, haze is expected to occur during this period. However, during the NE monsoon (established early November until the middle of March), steady easterly or north-easterly winds of 5.14 to $10.3\,\mathrm{m\,s^{-1}}$ prevail. During periods of strong surges of cold air from the north (cold surges), the winds over the east coast states of Peninsular Malaysia may reach $15.4\,\mathrm{m\,s^{-1}}$ or more. With the highest rainfall intensity and the possibility of flooding, NE monsoon is known as the wet season. In this study, air pollution episodes are defined considering $PM_{2.5}$ mass (more than $40\,\mathrm{\mu g\,m^{-3}}$) and the Air Pollution Index (API; more than 50), hereafter defined as HAZE samples. Local wind rose, seasonal regional synoptic wind field and biomass fire hotspots are given in Fig. S1 in the Supplement. The average temperature (T) at the site during the sampling campaign was $28.5 \pm 1.19\,^{\circ}\mathrm{C}$ and the average RH was $71.2 \pm 7.91\,\%$. Following the trend of T and API, WS was highest during the SW monsoon at an average of $1.39 \pm 0.187\,\mathrm{m\,s^{-1}}$ and lowest during the NE monsoon at $1.20 \pm 0.167\,\mathrm{m\,s^{-1}}$ with an annual average of $1.29 \pm 0.194\,\mathrm{m\,s^{-1}}$. Rainfall was lowest during the SW monsoon (6.27 ± 10.6 mm) and highest during the NE monsoon (15.1 ± 22.7 mm). Overall, the main wind direction (WD) for the site was south-easterly, that is east-south-east, south-east and south-south-east. Details of the meteorological and gaseous pollutants for each season are given in Table S1 in the Supplement.

2.2 Aerosol sampling

The aerosol sampling was conducted from 4 August 2011 to 17 July 2012, for 8 consecutive days every month (inclusive of one field blank) during a 1-year sampling period. Sampling (24 ± 1 h; around 09:00 to 09:00 LT, GMT+8) was performed using a high-volume $PM_{2.5}$ sampler (Tisch Environmental, Inc., USA; model TE-6070V-2.5-BL) running at $1.13\,\mathrm{m^3\,min^{-1}}$. Filter media used for sample collection were quartz micro-fibre filters (Whatman, United Kingdom; QMA catalogue number 1851–865) 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 $500\,^{\circ}\mathrm{C}$ for 3 h inside a furnace (Nabertherm, Germany; model L 5/11). 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 five-digit semi-micro analytical balance (A&D, USA; model GR-202) 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 $-18\,^{\circ}\mathrm{C}$ in a freezer prior to analysis.

2.3 Chemical analyses

2.3.1 Major ions

For the purpose of soluble ion analysis, one strip ($2.54\,\mathrm{cm} \times 20.32\,\mathrm{cm}$) of loaded quartz filter was used. The portion was cut into smaller pieces ($1\,\mathrm{cm} \times 1\,\mathrm{cm}$) directly into a 50 mL conical flask. Then 20 mL of ultra-pure water (UPW) (Hach USA; Millipore Direct-Q 3 UV System) with a resistivity of 18.2 MΩ were added and the flask capped with a stopper. For sonication extraction purposes ($60\,^{\circ}\mathrm{C}$; 60 m), an ultrasonic bath (Elma Schmidbauer GmbH, Germany; Elmasonic S40) was used. The solution was subsequently filtered through 0.2 µm 25 mm Acrodisc filters (Pall, USA; Part number 4612) using a $20\,\mathrm{cc\,mL^{-1}}$ 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 $4\,^{\circ}\mathrm{C}$ to allow for equilibrium of the solution 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 (MDLs) were calculated based on 3 times the standard deviation of field blank ($n = 6$) while 1 ppm standard of single-cation/anion standard (Certipur® reference materials for ion chromatography, Merck Millipore, Merck KGaA, Darmstadt, Germany) was used for the calculation of percentage recoveries. The percentage recoveries for all elements were between 86 and 131 %, as reported in 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 BV, Netherlands; MLS-1200 Mega). For the digestion process, one strip (2.54 cm × 20.32 cm) of loaded filter was used with the following settings of time (m) and power (W) were used: (1) 1, 250; (2) 1, 0; (3) 8, 250; (4) 5, 400; and (5) 5, 650. The solution was subsequently filtered through 0.2 µm 25 mm Acrodisc filters (Pall Gelmann) using a 50cc 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 (PerkinElmer Instrument, USA; model Elan 9000). MDL was estimated as 3 times the standard deviation of field blank ($n = 6$) while 1 ppm Multi-Element Calibration Standard 3 (PerkinElmer Pure Plus; PerkinElmer, USA) was use for validation purposes. Percentage recoveries are based on SRM1648a Urban PM (National Institute of Standards and Technology, MD, USA) and these varied between 29 and 101 %, as reported in Table S2. Details of experimental quality assurance and quality control for both trace elements and major ions are provided in the Supplement.

2.3.3 Black carbon

BC concentration was determined using a smoke stain reflectometer with calibration (Diffusion Systems Ltd., UK; model EEL 43M). In brief, this method involves the measurement of the darkness of the stain (on the filter paper) through its reflectance of white light (using a reflectometer). The reflectance is relative to the light reflected by a clean filter of the same material with an assumption of 100 % reflection. The absorbed light then converted (through calculation) for BC mass. In this study, 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; Moldanová et al., 2013).

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, Malaysia. The meteorological parameters included temperature (T), RH, WS, WD and daily values of API readings, while the gaseous pollutants were carbon monoxide (CO), ozone (O_3), sulfur dioxide (SO_2), nitrogen oxides (NO_x), nitrogen monoxide (NO) and nitrogen dioxide (NO_2). The instrument and measurement principle used for the gases were as follows: O_3 using Analyzer 400A (chemiluminescence); NO, NO_2, NO_x using Teledyne Advanced Pollution Instrumentation 200A (chemiluminescence); SO_2 using Teledyne Advanced Pollution Instrumentation M100A (fluorescence); and CO using Teledyne Advanced Pollution Instrumentation M300 (nondispersive infrared absorption). API for Malaysia is calculated based on five major air pollutants: 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} (particles with aerodynamic diameter less than 10 µm) 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, or using Microsoft® Excel 2010 with the statistical add-in XLSTAT version 2014.3.04. 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 data set (u, v-wind) downloaded from the National Center for Environmental Protection (NCEP)/National Center for Atmospheric Research (NCAR, http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.pressure.html, Kalnay et al., 1996; NOAA, 2015). The data set used was set at 500 mb with a mapping covering latitude of (−10°, 20° N) and longitude of (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 were used. Data were downloaded from the National Aeronautics and Space Administration (NASA) Land, Atmosphere Near real-time Capability for Earth Observing System (LANCE) Fire Information for Resource Management System (FIRMS) fire archive (https://firms.modaps.eosdis.nasa.gov/download/request.php) in the range of 10° S to 20° N and 90 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 500 m with 6 h trajectory intervals. 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, dust, secondary inorganic aerosol (SIA), trace element (TE), BC, K^+ and also the unidentified portion of the PM 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 used by Bressi et al. (2013). The dust fraction was calculated as the contribution of non-sea-salt Ca^{2+} in mineral dust. The 8.3 % mineral dust mass contribution for the Klang Valley area estimated by Rahman et al. (2011) was employed for the calculation. Following the direct CMC non-sea-salt Ca^{2+} approach, we therefore exclude the major mineral dust elements (Al, Fe) to calculate the rest of trace element mass contribution.

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

$$[\text{ss-}SO_4^{2-}] = 0.252 \times [Na^+]$$
$$[\text{dust}] = [\text{nss-}Ca^{2+}]/0.083$$
$$[\text{SIA}] = [\text{nss-}SO_4^{2-}] + [NO_3^-] + [NH_4^+],$$

with $[\text{nss-}SO_4^{2-}] = [SO_4^{2-}]–[\text{ss-}SO_4^{2-}]$, where ss stands for sea salt and nss stands for non-sea salt.

2.5.3 Enrichment factor (EF)

All elements that have high recovery were used for EF analysis. Due to the low recovery of Al, in this study we opted to use Fe as our reference element. Fe were also listed by Lawson and Winchester (1979) as reference for elemental EFs calculation besides Al, Si and Ti. Studies by Ho et al. (2006), Kuo et al. (2007) and Han et al. (2009) have successfully used Fe for their EF assessment. For the cut-off point, we follow Cesari et al. (2012). The study derived a two-threshold system of EF in which, for re-suspended soils, elements with an EF of smaller than 2 were considered to be from crustal sources, those with an EF of larger than 4 were considered from an anthropogenic origin, while those in between were considered of mixed origin.

2.5.4 Source apportionment

A combination of PMF version 5.0 (PMF 5.0) and MLR analysis was employed to determine source apportionment where results of the MLR were used to apportion the $PM_{2.5}$ chemical compositions in order to quantify sources. Details of the PMF procedure used in this study are similar to our previous work as discussed in Khan et al. (2015b). In brief, two data files were used as an input, i.e. (1) concentration and (2) uncertainty. For the concentration data file, the chemical composition data set were first pre-treated and validated. To ensure a strong signal from the data was evident, species with more than 50 % of the data below MDL were discarded. For the rest, the missing values were replaced by half of the MDL while data with values, but below MDL, were left as they were. The final data set used for the PMF analysis contained 80 samples with 31 elements (including $PM_{2.5}$ mass) whereby only elements that have high recovery were used. Based on the signal-to-noise (S / N) ratio, NO_3^- and Na^+ were set as “weak” species ($0.2 \leq S / N < 2$), while the rest was categorised as “strong” species (ratio ≥ 2; Heo et al., 2009; Richard et al., 2011; Yu et al., 2013). The $PM_{2.5}$ mass was also categorised as “weak” so as not to affect the PMF solution. The second data file is the uncertainty value of each variable in each sample estimated from an empirical equation. An additional 5 % uncertainty was added to account for methodological errors during preparation of filter papers, gravimetric mass measurements and preparing the calibration curves. Upon running the PMF analysis, different numbers of factors and F_{peak} values have been explored to obtain the most meaningful results with 100 bootstrap runs and a minimum R^2 of 0.6 to test the uncertainty of the resolved profiles. F_{peak} is a parameter uses to execute the test of rotational ambiguity and the bootstrap is used to detect and estimate the disproportionate effects or errors of a small set of observation resampling randomly. Paatero et al. (2014) suggested that each resampled version of observation, some randomly chosen rows of the original matrix occur multiple times, while other rows do not occur at all. Similar to base run, each resampled data set is decomposed into profile and contribution matrices using PMF. Our observations of each PMF run using the each starting point of seed from 1 until 9 are summarised in Table S3. The results showed that the solution was rather stable at the starting point of seed 9. It was observed that a five-factor solution provided the most meaningful results, based on the lowest Q (robust) and Q (true) value of 1581.27 with the $Q_{(\text{true})}/Q_{\text{exp}}$ value of 0.94 after 390 computational steps and the convergence of the results. PMF factors were resolved on 20 runs and seed value of 9, with $F_{\text{peak}} = 0$ found to be the most reasonable. The model output of source contribution is provided as normalised or dimensionless (average of each factor contribution is one). To express the output of PMF, the mass concentrations of the identified sources were scaled by using the MLR analysis.

Table 1. Descriptive statistics of $PM_{2.5}$ mass and particulate matter (PM) ratio; unit: mean ± standard deviation (min–max). Remarks: SW is the south-west monsoon; NE is the north-east monsoon; INT.2 is inter-monsoon 2; INT.1 is inter-monsoon 1; HAZE indicates samples with $PM_{2.5}$ mass more than $40\,\mu g\,m^{-3}$ and air pollution index (API) more than 50.

	ANNUAL	SW	INT.2	NE	INT.1	HAZE
Elements	5 Aug 2011–18 July 2012 $n = 81$	15 May–14 Sept $n = 29$	15 Sept–31 Oct $n = 7$	1 Nov–14 Mar $n = 35$	15 Mar–14 May $n = 10$	$n = 11$
$PM_{2.5}$ ($\mu g\,m^{-3}$)	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 ± 0.070
$PM_{2.5}$ / TSP	0.46 ± 0.13	0.50 ± 0.081	0.44 ± 0.12	0.40 ± 0.087	0.54 ± 0.22	0.54 ± 0.069
PM_{10} / TSP	0.63 ± 0.12	0.70 ± 0.087	0.71 ± 0.058	0.57 ± 0.12	0.65 ± 0.087	0.73 ± 0.12

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 (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 5 times the WHO daily guideline and more than 3 times the 24 h US EPA NAAQS standards. This value was recorded during the haze episode in June 2012. As shown in Fig. 2b, d and e, strong variability can be observed from the monthly and daily averages of $PM_{2.5}$ mass. 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 at $33\,\mu g\,m^{-3}$, while the lowest was on Wednesday ($24\,\mu g\,m^{-3}$). Meanwhile, weekends on average recorded lower $PM_{2.5}$ mass ($26\,\mu g\,m^{-3}$) compared to weekdays ($29\,\mu g\,m^{-3}$).

$PM_{2.5}$ mass shows significant variability between the NE monsoon and the three other seasons (SW, INT.2 and INT.1). Figure 2c showed that 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. The small number of exceedances during the NE monsoon was due to high rainfall (precipitation) 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 Asian cities (Reid et al., 2013). As shown in Fig. 2a, it is important to note that haze events always occur during the SW monsoon, and 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 $61 \pm 24\,\mu g\,m^{-3}$, higher compared to the 2011 haze episode documented for Bangi area at $48 \pm 10\,\mu g\,m^{-3}$ by Amil et al. (2014).

The annual $PM_{2.5}$ mass (weekly average representative of the month) for this study averaged at $28 \pm 18\,\mu g\,m^{-3}$. This is almost triple (2.8-fold) the $10\,\mu g\,m^{-3}$ WHO $PM_{2.5}$ annual guideline, 2.33-fold higher than the US EPA NAAQS $PM_{2.5}$ annual standard of $12\,\mu g\,m^{-3}$ and 1.12-fold higher than the European Union (EU) $PM_{2.5}$ annual standards set at $25\,\mu g\,m^{-3}$ (European Commission, 2015). Table 2 reports that $PM_{2.5}$ mass average for this study was very low compared to other big 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). 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

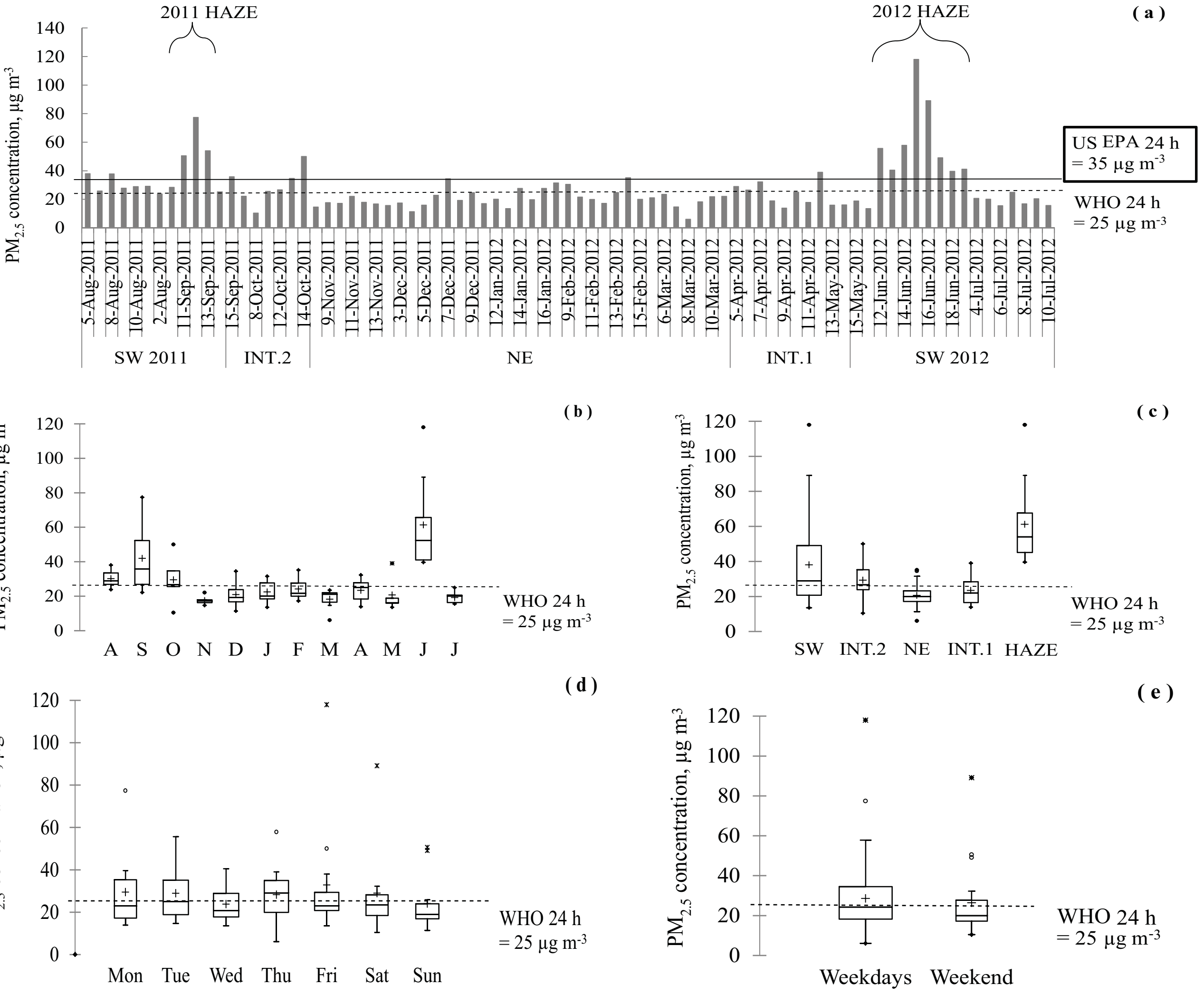

Figure 2. The $PM_{2.5}$ mass concentration on the **(a)** daily basis, with box and whisker plots of the **(b)** months, **(c)** seasons, **(d)** days of the week and **(e)** weekdays/weekends. For the box and whisker plots, the horizontal line within the box indicates the median, boundaries of the box indicate the 25th and 75th percentile and the whiskers indicate the highest and lowest values of the results. The "+" marked in the box indicates the mean. 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.

result for Petaling Jaya is 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, as reported in Table 1. PM_{10} / TSP ratio was 0.63 ± 0.12. The $PM_{2.5}$ / PM_{10} ratio at this site was higher than other studies in Asia as reported by Hopke et al. (2008), where most of the sites studied showed ratios of lower than 0.50. 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 0.70. Our $PM_{2.5}$ / PM_{10} ratio was also in agreement with other cities in Europe (Gehrig and Buchmann, 2003; Gomišček et al., 2004; Contini et al., 2014). Despite having different characteristics, the SW and NE monsoons still came out with similar values to the annual $PM_{2.5}$ / PM_{10} ratio at 0.72 ± 0.10 and 0.71 ± 0.13 respectively. The similar $PM_{2.5}$ to PM_{10} ratio during the wet and the dry season indicates that meteorological parameters, specifically rainfall, are affecting the fine (particle with an aerodynamic diameter of less than 2.5 µm) and coarse (particle with an aerodynamic diameter of greater than 2.5 µm) particles in the same way. This is also confirmed by the good correlation of $PM_{2.5}$ and PM_{10} ($r = 0.963$; $p < 0.0001$). 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)

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 July 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, suburban	Aug 2006–Dec 2007	Tahir et al. (2013b)
Petaling Jaya, Klang Valley, Malaysia Gombak, Klang Valley, Malaysia	33 28	Urban industrial Urban residential	Dec 1998–Dec 2000 Dec 1998–Dec 2000	Keywood et al. (2003)
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 Sept 2009–10 Sept 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, United Kingdom	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)

than INT.1 (0.85 ± 0.40). 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 ± 0.070. To further examine the particle at the site, the seasonal $PM_{2.5}$ / TSP ratio was calculated. During the dry season (the SW monsoon), ambient air at the site had particles in the ratio of approximately 50/50 coarse to fine particles ($PM_{2.5}$ / TSP $= 0.50 \pm 0.081$). 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 0.40 ± 0.087 respectively. INT.1 and HAZE episodes, in contrast, 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).

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

Referring to Table 3, the Pearson correlation revealed that $PM_{2.5}$ mass on an annual basis was significantly influenced by meteorological and gaseous parameters. 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 ($r = 0.310$; $p = 0.005$) 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 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, in contrast, showed a significant positive correlation with $PM_{2.5}$ mass at $r = 0.298$ ($p < 0.01$). The significant positive correlation of $PM_{2.5}$ and O_3 possibly indicates a secondary source of $PM_{2.5}$ as well as the already identified combustion-related traffic source, which is primary.

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 mon-

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 is temperature, RH is relative humidity, WS is wind speed and WD is wind direction.

(a)	Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
	API	**0.763**$^{\mathrm{b}}$	**0.748**$^{\mathrm{b}}$	0.299	**0.473**$^{\mathrm{a}}$	**0.705**	0.531
	T	**0.310**	0.236	0.572	0.201	0.030	−0.050
	RH	**−0.314**$^{\mathrm{a}}$	−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)							
	CO	**0.471**$^{\mathrm{b}}$	**0.687**$^{\mathrm{b}}$	0.713	**0.488**$^{\mathrm{a}}$	0.654	**0.749**$^{\mathrm{a}}$
	O_3	**0.298**$^{\mathrm{a}}$	**0.535**$^{\mathrm{a}}$	0.427	0.433	0.378	0.449
	SO_2	**0.324**	0.141	−0.250	**0.654**$^{\mathrm{b}}$	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**$^{\mathrm{b}}$	**0.528**$^{\mathrm{a}}$	**0.851**	**0.711**$^{\mathrm{b}}$	**0.874**$^{\mathrm{a}}$	0.599

Values in bold are different from 0 with a significance level alpha = 0.05; $^{\mathrm{a}}$ is when p values < 0.001 and $^{\mathrm{b}}$ p values < 0.0001.

soon, which is 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 show positive correlations. HAZE events revealed a slight negative correlation between $PM_{2.5}$ mass and T. This condition is perhaps because during haze episodes, the small particles envelope the atmosphere and reduce the UV radiation which can reduce the temperature of earth surface. RH and $PM_{2.5}$ mass, however, revealed negative relationships with three seasons (except INT.1) having low correlations. INT.1 showed the reverse relationship. However, HAZE events which occur during the SW monsoon disagree 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, Table 1 reported that the $PM_{2.5}$ / PM_{10} ratio for both major seasons (SW and NE) were almost the same at ~ 0.70. The $PM_{2.5}$ / TSP and PM_{10} / TSP ratios were different, however. During the SW monsoon ratios of 0.50 and 0.70 were observed, while during the NE monsoon ratios of 0.40 and 0.57 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 greatly influence only the coarse particles (PM $dp > 2.5\,\mu m$) 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 % ($4.15\,\mu g\,m^{-3}$) of the $PM_{2.5}$ mass. The total anion mass measured was $1.67\,\mu g\,m^{-3}$ (6.0 % of $PM_{2.5}$ mass) while the total cation mass was $1.75\,\mu g\,m^{-3}$ (6.3 % of $PM_{2.5}$ mass). As shown in Fig. S2, the equivalent charge ratio of total cation to total anion ratio was 0.46 indicates that the aerosol at the site is acidic due to the excess of anions, also experienced by another study (He et al., 2012). 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 ions; 23 % of IM mass) > NH_4^+ (29 % of water-soluble ions; 17 % of IM mass) > Na^+ (9 % of water-soluble ions, 5 % of IM mass) > K^+ (7 % of water-soluble ions; 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, contrastingly, accounted for about 8.6 % of $PM_{2.5}$ mass ($2.41\,\mu g\,m^{-3}$) 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 > 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 % trace element recorded at Kuala Lumpur city (Rahman et al., 2011) but higher compared to Kuala Terengganu (Tahir et al., 2013b).

3.3 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.

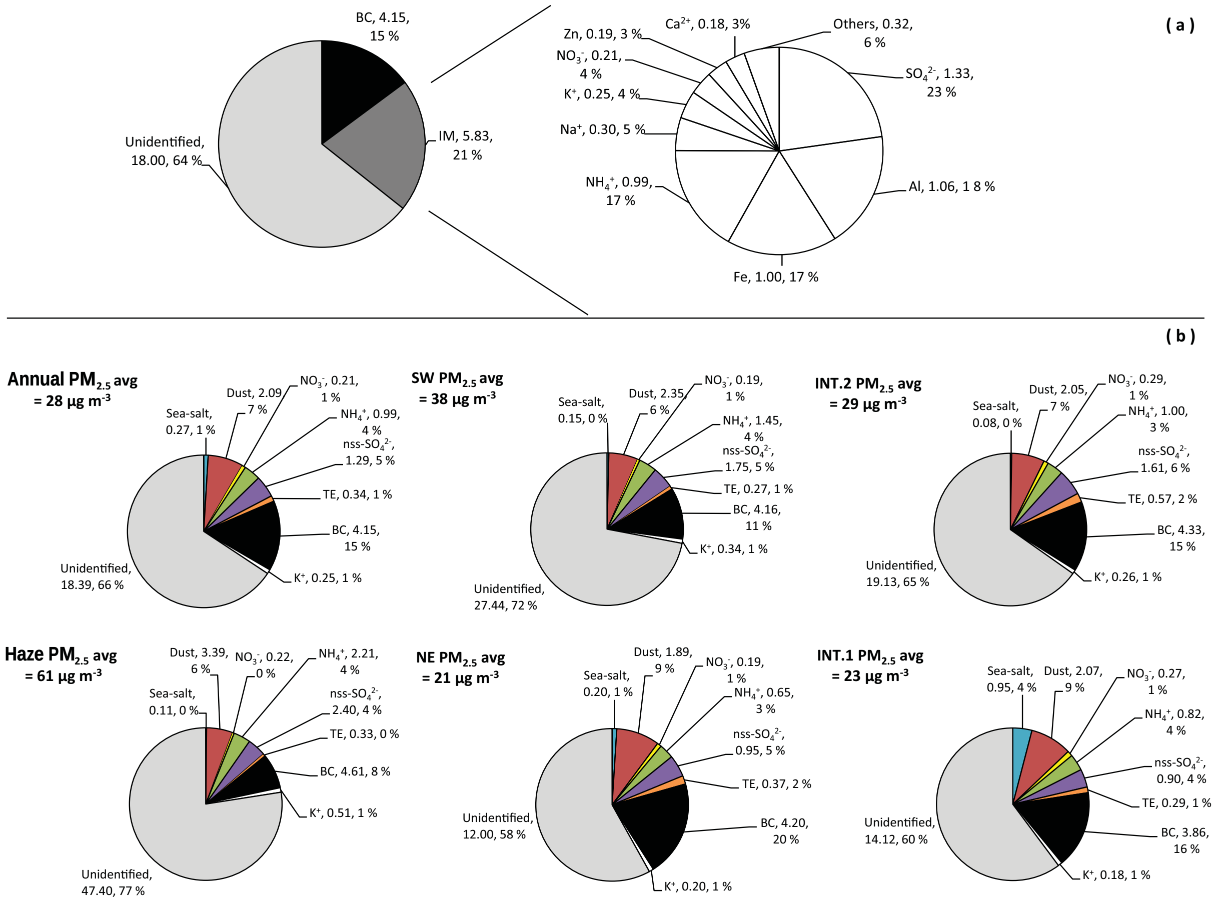

Figure 3. The composition of $PM_{2.5}$ displayed as (element, mass in $\mu g\,m^{-3}$, percentage in $PM_{2.5}$ mass) based on **(a)** annual chemical composition determined where IM is the inorganic matter and **(b)** seasonal chemical mass closure (CMC) components identified.

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 %. Therefore, 66 % was left unidentified which was presumed to be sulfur (S) compounds 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 ($0.344\,\mu g\,m^{-3}$; 1 %) > sea salt ($0.265\,\mu g\,m^{-3}$; 1 %) > K^+ ($0.253\,\mu g\,m^{-3}$; 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 and 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 = $0.987\,\mu g\,m^{-3}$; 4 % of $PM_{2.5}$ mass; 17 % of IM mass) and NO_3^- ($0.213\,\mu g\,m^{-3}$; 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 to 2008 by Keywood et al. (2003) at 98 % for both nss-SO_4^{2-} and nss-K^+, which is why 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, as reported in Table S1. The NR was calculated based on the ratio of the NH_4^+ (eq m^{-3}) to the sum of SO_4^{2-} and NO_3^- (eq m^{-3}) (Squizzato et al., 2013). The overall NR obtained for this site was 0.26, indicating an excess of SO_4^{2-} and NO_3^-. The NR ratio varied with season. The highest recorded NR was during the HAZE episodes with 0.35. The rest of the values showed the following trend: SW (0.31) > NE (0.22) > INT.2 (0.21) > INT.1 (0.17).

Trace elements, which are good indicators for anthropogenic factors, had a mass contribution of 0.344 µg 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, 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 ($EF \geq 1000$): 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 several elements (Cu, Rb, V and Ni), as shown in Fig. S3. For example, Cu during SW monsoon follows the annual grouping of anthropogenic source while, during other seasons, it is drawn from the crustal source. Meanwhile, Rb, V and Ni during the SW monsoon originate from the anthropogenic source, which is contrary to the annual and other seasonal patterns. Ni and V are known as heavy oil combustion indicators (Jiang et al., 2014), Cu is known to be associated with the traffic (Contini et al., 2014), while Rb is known to be drawn from the crustal source (Khan et al., 2010a). A study in Taiwan also argued that these four elements (Cu, Rb, V and Ni) are likely to be affected by both soil and non-soil emissions (Balakrishnaiah et al., 2012).

Dust was 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 Southeast 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 Sumatra and Jawa Island), as shown in 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 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. (2010b) 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 (0.253 ± 0.144 µg m^{-3} on annual scale) regardless of the season change, including the HAZE episodes.

BC averaged at 4.15 ± 0.642 µg 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-monsoon 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 µg m^{-3} EC with 74 µg m^{-3} of organic carbon (OC) in TSP samples (TSP mass of 300 µg m^{-3}) during haze episodes, while during normal days they found 8 and 14 µg m^{-3} for EC and OC respectively from 74 µg 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 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 low compared to most other SEA cities as reported by Reid et al. (2013). One possible reason is because this study was carried out on a long-term basis while the others mostly concentrate on a particular season and/or event, especially haze episodes.

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 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 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.

3.4 Source apportionment and its relation to meteorological–gaseous conditions

Referring to Table 4 and Fig. 4a, 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. 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 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).

3.4.1 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).

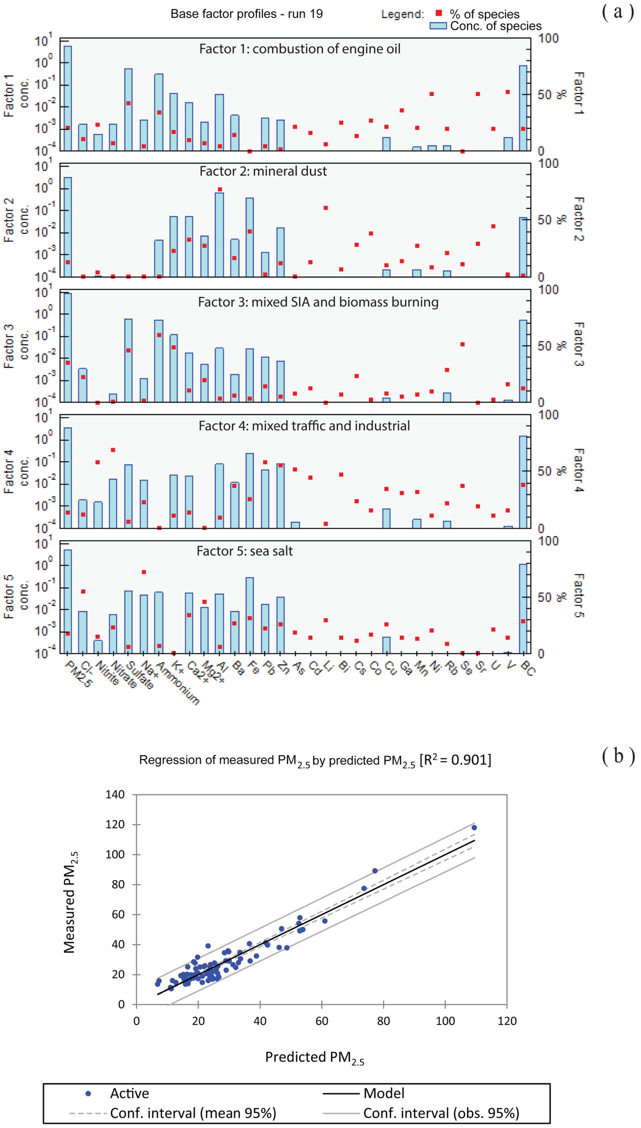

Figure 4. Source apportionment results from positive matrix factorisation (PMF) analysis: **(a)** source profile and **(b)** regression plot between measured and predicted PM$_{2.5}$ mass (both in µg m^{-3}). Remark: SIA is the secondary inorganic aerosol.

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 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

Table 4. Relative contribution of $PM_{2.5}$ sources from the positive matrix factorisation (PMF) analysis. Remarks: SIA is the 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 %)	11.3 (19 %)
Factor 3: mixed SIA and biomass burning	11.7 (42 %)	19.1 (51 %)	9.99 (35 %)	7.44 (34 %)	6.21 (26 %)	36.9 (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 %)	10.8 (45 %)	4.62 (8 %)

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, as reported in Table S4. 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 occurred during SW ($r = 0.581$; $p = 0.007$), which may have resulted from HAZE ($r = 0.677$; $p = 0.045$). For gaseous parameters, factor 1 seemed to correlate with 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. The positive correlation during INT.2 was perhaps due to higher measurements of NO_x and NO_2 during this time period compared to other seasons. NO_2 provides an O radical which contributes to the formation of O_3 with the assistance of sunlight and volatile organic compounds (VOCs). High concentrations of O_3 and other organic pollutants can lead to the formation of secondary organic aerosol; this may explain the observation results.

3.4.2 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) and 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, in contrast, 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. This factor during HAZE seems to be affected by a few gaseous parameters, i.e. 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 $r = 0.260$ ($p = 0.035$). 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 record 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.

3.4.3 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^+), however, is an indication of major soil elements, usually from biomass burning. Echalar

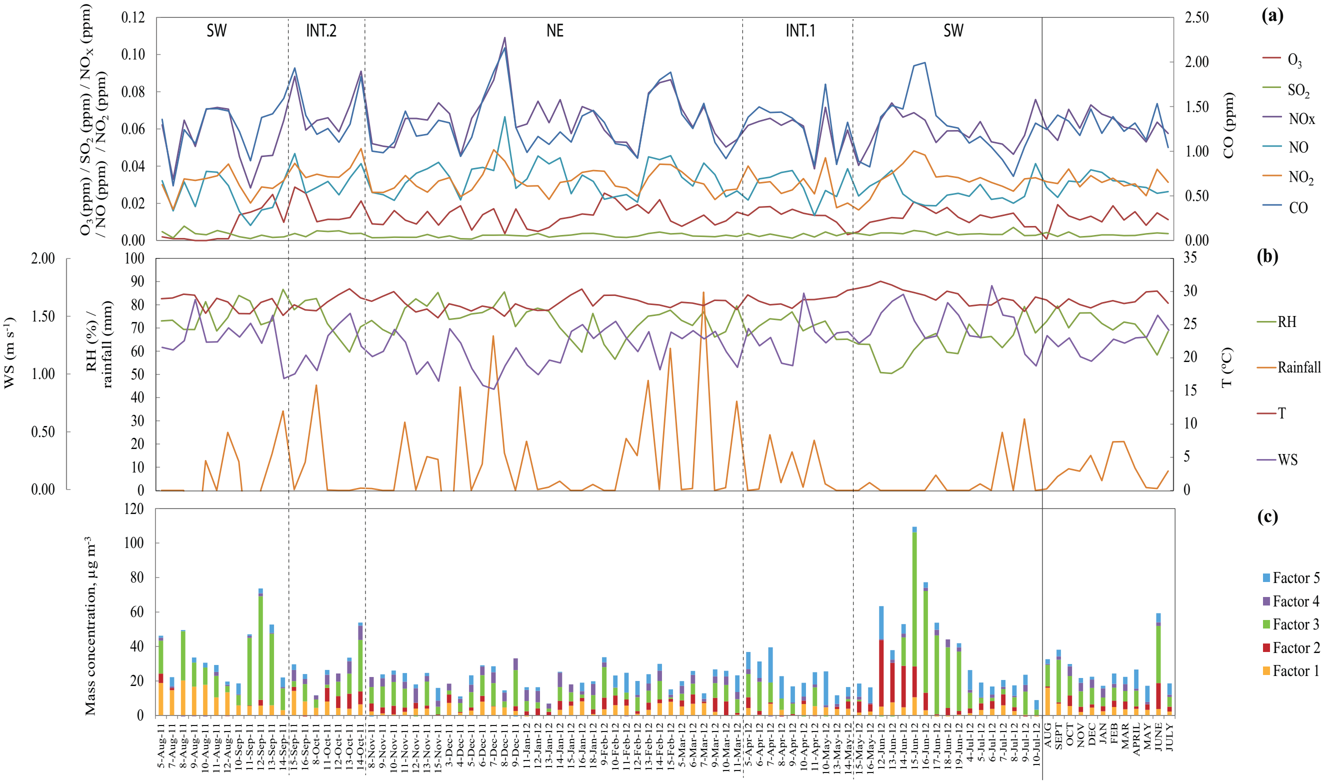

Figure 5. Time series of daily and monthly variations (left to right) of **(a)** gaseous, **(b)** meteorological parameters and **(c)** mass concentration of $PM_{2.5}$ sources.

et al. (1995) have 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, K^+ in PM was seen in many studies as a marker of biomass origin, in either the European region (Reisen et al., 2013) or the 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. Characterised by high levels of NH_4^+ (59 % of NH_4^+ 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. (2006) 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, represents 35 % or less of the $PM_{2.5}$ mass, i.e. 35, 34 and 26 % respectively. Except for INT.1, the other seasons show very significant correlations between this factor and secondary aerosol components, i.e. SO_4^{2-}, NH_4^+ and K^+. During HAZE, this factor contributed 63 % of the $PM_{2.5}$ mass. The time series (Fig. 5c) shows that this factor's elevated contribution occurred during a period from June until the end of October, which is when the haze episodes normally occur. The HYSPLIT back-trajectory analysis traced back the mass from the HAZE samples to Sumatra, i.e. Palembang during the 2011 episode and Palembang/Pekan Baru for 2012 episode; see Fig. S4a(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 factor 3 while NO revealed a significant negative correlation. The strongest correlations between this factor and meteorological–gaseous parameters were observed during the SW monsoon season. With highest mass contribution during SW, this factor showed 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, however, although normally occurring during the SW monsoon, did not share these relationships. This factor during HAZE only correlated strongly with WS ($r = -0.678$; $p < 0.05$).

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

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 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 industrial emissions (Song et al., 2006; Wåhlin et al., 2006; Querol et al., 2008; Pey et al., 2009; Thurston et al., 2011; Srimuruganandam and Shiva Nagendra, 2012a, b). 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 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 such as motor vehicle exhaust, industries (petrochemical industry, iron/steel plant, etc.) and stationery combustion sources (such as coal plants).

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, this factor during 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. During the NE dry season, this factor showed a negative 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 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.

3.4.5 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 Table 4 and the time series illustration of Fig. 5c, 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 = 0.10 , INT.2 = 0.083, NE = 0.072 and INT.1 = 0.24. The Cl^- / Na^+ ratios for all seasons were SW = 0.11, INT.2 = 0.056, NE = 0.14 and INT.1 = 0.041, with an overall annual ratio of 0.057. From these results, it is obvious that INT.1 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. (2010b) 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 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 $r = -0.816$ ($p = 0.048$).

3.4.6 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, as reported in Table S5, revealed the two episodes to have quite different characteristics. 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. For a total of 19 % during HAZE 2011, four other factors were identified: combustion of engine oil, sea salt, mineral dust and mixed traffic and industrial. These factors do not seem to have a strong influence on HAZE 2011. However, HAZE 2012 was strongly influenced by those four factors, with a combined contribution of 44 %. As reported in Table S6, 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. 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-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 Sumatra, Indonesia; see Fig. S4a(ii), b(ii). Further analysis showed that HAZE 2012 was more influenced by the meteorological and gaseous parameters, whereas during HAZE 2011, strong correlations were observed but they are not significant, as shown in Table S7. However, it is still not clear whether long-range transport did impact our HAZE episodes.

3.5 Comparison between CMC and PMF Source

As shown in Figs. 4b and S5, predicted mass modelled by PMF and reconstructed mass by CMC were compared to those measured $PM_{2.5}$ mass. Both approaches resulted with good regression at $R^2 = 0.901$ and $R^2 = 0.784$ respectively. Further, seasonal regressions and time series between these two approaches were shown in Fig. S6. The analysis were run on selected components that have similarity to compare (1) CMC dust vs. PMF factor 2 mineral dust; (2) CMC SIA and K^+ vs. PMF factor 3 SIA and biomass burning; and (3) CMC sea salt vs. PMF factor 5 sea salt.

Overall, "dust" and "SIA and biomass" components show similar trend, as shown in Fig. S6. Both components have good correlations between CMC and PMF approach (R^2 more than 0.70) except during INT.1 and NE monsoon (R^2 less than 0.50). Rainfall, which was higher during these two seasons compared to other seasons, could be the reason. The ANNUAL CMC/PMF ratio for seasonal dust component is 0.29 ($r = 0.89$) while seasonal regression (including HAZE) ranges between 0.24 and 0.53. The seasonal CMC / PMF ratio for the SIA and biomass component ranges between 0.13 and 0.24 with an annual ratio of 0.15 ($r = 0.87$). The inconsistency ratio for both components maybe due to the fact that PMF contribution includes absorbed elements other than selected elements considered by the CMC approach. The "sea salt" component, however, did not show good agreement between two approaches. However, for HAZE data set, the two approaches seem to have a very good agreement ($r = 0.94$) for the sea salt component. As shown in Fig. S6c, the seasonal regression of CMC and PMF for sea salt showed that this pair is moderately correlated. Similar observation of large difference for the sea salt component (also known as marine aerosol) between two approaches with one of them being the mass closure calculation seen and discussed by Almeida et al. (2006) and Farao et al. (2014). The observation on this matter was that perhaps CMC calculation did not include all the components associated with sea spray and the reaction of NaCl with inorganic acids (HNO_3 and H_2SO_4) resulted in the loss of Cl^- ion.

The different estimation derived from the two approaches was expected. According to Harrison et al. (2003), CMC is a hybrid between comprehensive chemical analysis method and simpler statistical procedure. It is a simple approach yet an effective model to assort the measured PM compounds into different source categories. One of the highlights of this method is that CMC treats sulfate and nitrate separately. This is crucial since different ambient conditions can lead to different responses of the aerosol, which will further affect organic carbon and thus secondary organic carbon trend. Moreover, CMC also separates the sea salt and crustal components which have different responses to changes in traffic volume. PMF, in contrast, is an advanced computational tool to identify sources and eventually the mass contribution based on the work by Paatero and Tapper (1994). So, it is likely there are two different results and thus it is almost impossible to verify results from the two different methods (Hellén et al., 2003; Hopke et al., 2006; Vallius et al., 2008; Vecchi et al., 2008; Favez et al., 2010; Hellebust et al., 2010). These issues have been highlighted by Viana et al. (2008) regarding European source apportionment studies. The study stated that it is difficult to obtain coinciding results with different receptor models for the same data. This statement is supported by Vallius et al. (2008) who stated that different methods yield different results when they are applied to air pollution data.

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 and $118\,\mu g\,m^{-3}$ with 43 % (samples) exceedance of the daily WHO guideline. On average, weekends recorded lower $PM_{2.5}$ mass ($26\,\mu g\,m^{-3}$) compared to weekdays ($29\,\mu g\,m^{-3}$). The month of June during the dry season of the SW monsoon recorded the highest monthly average at $61\,\mu g\,m^{-3}$ while November during the wet season of the NE monsoon recorded the lowest ($17\,\mu g\,m^{-3}$). 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 the 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 the 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 (particles with an aerodynamic diameter of greater than 2.5 µm) but not so much the fine particles (particles with an aerodynamic diameter of less than 2.5 µm) at this site.

The determined $PM_{2.5}$ chemical compositions 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 following 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 of 0.26 indicated an excess of SO_4^{2-} and NO_3^- at the site. 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 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 Sumatra, 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. However, correlation between the chemical constituents and sources of $PM_{2.5}$ towards meteorological and/or gaseous parameters largely varied with different season. Overall, this study suggests that $PM_{2.5}$ and its constituents here in Klang Valley urban-industrial environment were characterised by the local and regional activities as well as the seasonal tropical change. However, our study is limited to only fine particles. Parallel sampling of both fine and coarse particles will give better insight into the actual condition of the aerosol at a site. With the use of meteorological–gaseous parameters, concrete conclusion can be achieved as to whether meteorological–gaseous parameters affect in the same way fine and coarse particles and whether both fine and coarse particles share common sources. In addition to inorganic composition, further comprehensive assessment covering the organic portion and total elemental inorganic composition (i.e. total K, total Mg, total Na, total Ca, Si, S) is necessary for a complete composition data set. In addition, it is suggested that particle number concentration distribution should be incorporated into the chemical composition SA analysis as well. The potential source contribution function could also enhance the analysis of local and regional long-range transport. Alternatively, a simple yet effective approach, that is looking at the prevailing wind direction (or polar plots) associated to the different PMF factors, could answer the effect of wind direction on PM. This would lead to improved analysis results and interpretation of the $PM_{2.5}$ data set, which eventually will lead to better understanding of the fine particle variability here in Klang Valley.

Data availability

Data sets for u-wind and v-wind used for the synoptic wind fields are publically available. These NCEP Reanalysis data were provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their website at http://www.esrl.noaa.gov/psd/.

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