

# 1 Seasonal Variability of PM<sub>2.5</sub> Composition and Sources in 2 the Klang Valley Urban-Industrial Environment

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## 17 18 **Abstract**

19 This study investigates the fine particulate matter (PM<sub>2.5</sub>) variability in the Klang Valley  
20 urban-industrial environment. In total, 94 daily PM<sub>2.5</sub> samples were collected during a one-  
21 year campaign from August 2011 to July 2012. This is the first paper on PM<sub>2.5</sub> mass, chemical  
22 composition and sources in the tropical environment of Southeast Asia, covering all four  
23 seasons (distinguished by the wind flow patterns) including haze events. The samples were  
24 analysed for various inorganic components and black carbon. The chemical compositions  
25 were statistically analysed and the temporal aerosol pattern (seasonal) was characterised using  
26 descriptive analysis, correlation matrices, enrichment factors (EF), stoichiometric analysis and  
27 chemical mass closure (CMC). For source apportionment purposes, a combination of positive  
28 matrix factorisation (PMF) and multi-linear regression (MLR) was employed. Further,

1 meteorological-gaseous parameters were incorporated into each analysis for improved  
2 assessment. In addition, secondary data of total suspended particulate (TSP) and coarse  
3 particulate matter (PM<sub>10</sub>) sampled at the same location and time with this study (collected by  
4 Malaysian Meteorological Department) were used for PM ratio assessment. The results  
5 showed that PM<sub>2.5</sub> mass averaged at  $28 \pm 18 \mu\text{g m}^{-3}$ , 2.8-fold higher than the World Health  
6 Organisation (WHO) annual guideline. On a daily basis, the PM<sub>2.5</sub> mass ranged between 6 and  
7  $118 \mu\text{g m}^{-3}$  with the daily WHO guideline exceeded 43% of the time. The North-east  
8 monsoon (NE) was the only season with less than 50% sample exceedance of the daily WHO  
9 guideline. On an annual scale, PM<sub>2.5</sub> mass correlated positively with temperature (T) and wind  
10 speed (WS) but negatively with relative humidity (RH). With the exception of NO<sub>x</sub>, the gases  
11 analysed (CO, NO<sub>2</sub>, NO and SO<sub>2</sub>) were found to significantly influence the PM<sub>2.5</sub> mass.  
12 Seasonal variability unexpectedly showed that rainfall, WS and wind direction (WD) did not  
13 significantly correlate with PM<sub>2.5</sub> mass. Further analysis on the PM<sub>2.5</sub>/PM<sub>10</sub>, PM<sub>2.5</sub>/TSP and  
14 PM<sub>10</sub>/TSP ratios reveal that meteorological parameters only greatly influenced the coarse  
15 particles (particles with an aerodynamic diameter of greater than 2.5  $\mu\text{m}$ ) and less so the fine  
16 particles at the site. Chemical composition showed that both primary and secondary pollutants  
17 of PM<sub>μ</sub> are equally important, albeit with seasonal variability. The CMC components  
18 identified were in the decreasing order of (mass contribution): black carbon (BC) > secondary  
19 inorganic aerosols (SIA) > dust > trace elements (TE) > sea salt > K<sup>+</sup>. The EF analysis  
20 distinguished two groups of trace elements: those with anthropogenic sources (Pb, Se, Zn, Cd,  
21 As, Bi, Ba, Cu, Rb, V and Ni) and those with a crustal source (Sr, Mn, Co and Li). The five  
22 identified factors resulting from PMF 5.0 were: 1) combustion of engine oil; 2) mineral dust;  
23 3) mixed SIA and biomass burning; 4) mixed traffic and industrial; and 5) sea salt. Each of  
24 these sources had an annual mean contribution of 17, 14, 42, 10 and 17%, respectively. The  
25 dominance of each identified source largely varied with changing season and a few factors  
26 were in agreement with the CMC, EF and stoichiometric analysis, accordingly. In relation to  
27 meteorological-gaseous parameters, PM<sub>2.5</sub> sources were influenced by different parameters  
28 during different seasons. In addition, two air pollution episodes (HAZE) revealed the  
29 influence of local and/or regional sources. Overall, our study clearly suggests that the  
30 chemical constituents and sources of PM<sub>2.5</sub> were greatly influenced and characterised by  
31 meteorological and gaseous parameters which largely vary with season.

32

## 1 **1 Introduction**

2 Airborne particulate matter (PM) significantly impacts global climate (Jacobson, 2002; Vieno  
3 et al., 2014; Mallet et al., 2016), causing visibility degradation in both urban and less polluted  
4 environments (Diederer et al., 1985; Doyle and Dorling, 2002; Watson, 2002; Chang et al.,  
5 2009; Hyslop, 2009) and accelerates material decay (Grossi and Brimblecombe, 2002). Fuzzi  
6 et al. (2015) revealed that climate-aerosol interaction, as well as effects of PM on human  
7 health and the environment, were underpinned by many new processes and development in  
8 the science. Different sizes of PM have been found to have varying toxicities impacting  
9 human health (Schwartz et al., 1996; Katsouyanni et al., 1997; Pope III, 2000; Ruuskanen et  
10 al., 2001; Eatough et al., 2003; Halonen, 2009; Ross et al., 2013; Khan et al., 2016). The  
11 fine particles, which are composed of compounds of a range of volatilities, appear to do more  
12 harm to human health than coarse particles (Dockery et al., 1993; Schwartz et al., 1996;  
13 Laden et al., 2000; Lanki et al., 2006; Pope III and Dockery, 2006; Krewski et al., 2009;  
14 Tagaris et al., 2009; WHO, 2013).

15 The fraction and composition variability of fine particles ( $PM_{2.5}$ ; particles with an  
16 aerodynamic diameter of less than  $2.5 \mu m$ ) are strongly influenced by seasonal  
17 meteorological factors, gaseous parameters and location. Megaritis et al. (2014) showed that  
18  $PM_{2.5}$  in Europe appears to be more sensitive to temperature changes compared to other  
19 meteorological and gaseous parameters in all seasons. Aside from meteorological and gaseous  
20 pollutants, seasonal changes and the background of an area (topography and local activities  
21 affecting anthropogenic and/or natural air pollution emissions) also influenced the  $PM_{2.5}$   
22 chemical variability (Tai et al., 2010; Tai et al., 2012). Seasonal variation of  $PM_{2.5}$  mass and  
23 its chemical composition for the Asian region has been widely reported. For example,  
24 Balasubramanian et al. (2003) reported that Singapore  $PM_{2.5}$  mass temporal variability was  
25 influenced by a number of factors including changes in emission strength, wind direction  
26 (WD) and other meteorological parameters. Also, their chemical mass closure (CMC)  
27 components (i.e. soil dust, metallurgical industry, biomass burning and automobiles, sea salt,  
28 and fuel oil combustion) at times were significantly attributed to Indonesian forest fires  
29 compared to local traffic and industrial emissions. Ye et al. (2003) reported varied CMC  
30 elements (ammonium sulfate and nitrate, carbonaceous material, crustal components,  
31 potassium) for Shanghai seasons where significant changes in the  $PM_{2.5}$  mass were observed  
32 with changing season. Meanwhile, sources of  $PM_{2.5}$  in Beijing (dust, secondary sulfate,

1 secondary nitrate, coal combustion, diesel and gasoline exhaust, secondary ammonium,  
2 biomass aerosol, cigarette smoke, vegetative detritus) showed distinct seasonal trends (Zheng  
3 et al., 2005). India PM<sub>2.5</sub> sources (i.e. motor vehicles, biomass burning, marine aerosol, tyre  
4 and brake wear, soil, secondary PM, and others) were observed to have considerable seasonal  
5 and weekday/weekend variations (Srimuruganandam and Shiva Nagendra, 2012b). A study by  
6 Louie et al. (2005) on PM<sub>2.5</sub> chemical compositions showed variations between different  
7 locations in Hong Kong where elevated concentrations of a source marker species at a site  
8 explained a higher influence of that source. The study identified carbonaceous aerosol as the  
9 largest contributor, followed by ammonium sulfate, crustal material, sea salt, and ammonium  
10 nitrate. Similar observations were also evident for Indonesia where source apportionment  
11 analysis on the elemental composition of PM revealed different numbers of factors for urban  
12 and suburban areas (Santoso et al., 2008).

13 PM<sub>2.5</sub> in the atmosphere consists of primary and secondary pollutants including volatile, non-  
14 volatile and semi-volatile components which originate from various sources (Eatough et al.,  
15 2006). Source apportionment (SA) is an approach that aims to identify and quantify the  
16 various sources of air pollutants (Hopke and Song, 1997; Watson et al., 2002; Wagstrom and  
17 Pandis, 2011). The most common method is receptor modelling. Receptor modelling  
18 measures atmospheric concentrations of chemically-speciated particles to infer the sources  
19 responsible for their emission, or the pathways of formation of secondary particles (Viana et  
20 al., 2008). The method starts by collecting and measuring ambient PM at a receptor (location),  
21 and works backwards to determine the sources. Receptor modelling uses temporal and  
22 chemical variations to separate total PM into different factors, where marker species are used  
23 to identify the sources. The goal of receptor models is to solve the chemical mass balance  
24 between measured species concentrations and source profiles. One of the models used to solve  
25 the chemical mixture is positive matrix factorisation (PMF), first developed by Paatero and  
26 Tapper (1993). Subsequently, numerous other studies have employed this method into their  
27 PM<sub>2.5</sub> receptor modelling including many undertaken in the Asian region. For example,  
28 Begum et al. (2004) have successfully applied PMF on inorganic and BC datasets to lead to  
29 source identification for PM<sub>2.5</sub> in Bangladesh. Srimuruganandam and Shiva Nagendra (2012b)  
30 made an evaluation of PM<sub>2.5</sub> sources for Chennai city, India using only inorganic (elemental)  
31 compositions. A study by Zhang et al. (2013) has successfully discussed the seasonal  
32 perspective of PM<sub>2.5</sub> sources (soil dust, coal combustion, biomass burning, traffic and waste

1 incineration emissions, industrial pollution, secondary inorganic aerosol) in Beijing, China  
2 using PMF on inorganic and organic datasets. Similar applications of PMF to apportion the  
3 sources of  $PM_{2.5}$  have also been successfully carried out here in Southeast Asia (SEA). For  
4 example, Santoso et al. (2008) used inorganic and BC datasets to identify five major sources  
5 of  $PM_{2.5}$  as biomass burning, soil, two stroke engine emissions, sea salt, secondary sulfate,  
6 motor vehicle emissions, and road dust. A study by Rahman et al. (2011) also used similar  
7 chemical compositions for the SA analysis of  $PM_{2.5}$  samples from the Klang Valley, which  
8 resulted in five sources: two stroke engine emissions, motor vehicle emissions,  
9 smoke/biomass burning, soil and industry. PMF was also effectively applied by Khan et al.  
10 (2015b) to their polycyclic aromatic hydrocarbons (PAHs) dataset to characterise the  $PM_{2.5}$  for  
11 the semi-urban area of Bangi, Malaysia. This study revealed three main sources: gasoline  
12 combustion, diesel and heavy oil combustion, and natural gas and coal burning. One of the  
13 current trends of SA is to apply more than one receptor model, a trend set by a number of  
14 countries i.e. Belgium, Germany, Portugal and Spain (Viana et al., 2008). Due to limitations  
15 of a single model, applying more than one receptor model will enhance the SA analysis,  
16 leading to enhanced characterisation of an element and/or source and thus increasing the  
17 confidence in interpretations from the results. The study also reports that the most frequent  
18 combinations used for SA are principal component analysis (PCA)-cluster analysis (CA),  
19 PCA-Lenschow, PCA-chemical mass balance (CMB), PCA-back-trajectory analysis, PMF-  
20 UNMIX-multilinear engine (ME), and CMB-mass balance.

21 Reid et al. (2013) discussed in detail how the SEA region holds a complex relationship  
22 between geographic, socio-economic, meteorological, and aerosol microphysical factors. The  
23 review emphasised timing and location of sampling when trying to achieve a representation of  
24 the actual condition of the aerosol system, as the urban and industrial aerosol environments  
25 differ between urban centres. For example, in Jakarta of Indonesia, two stroke engine  
26 vehicles, high emitters of particles and incomplete combustion products, were the major  
27 factor. Meanwhile, mobile sources are significant in Bangkok, Thailand, whereas Manila of  
28 Philippines was significantly affected by diesel truck and bus emissions. Having said that,  
29 most urban centres in the region share the major sources of meat cooking and oil-gas-  
30 petrochemical industry activity as well as shipping influences. In addition, the region is also  
31 affected by haze episodes caused by biomass burning. Taking this into consideration, we  
32 conducted a one-year assessment of  $PM_{2.5}$  covering all four seasons (including haze events) to

1 investigate its variability in the Klang Valley (urban-industrial) tropical environment. The  
2 samples were subjected to chemical measurements of inorganic matter (IM) compositions and  
3 black carbon (BC). We identified and apportioned the sources to PM<sub>2.5</sub> mass by employing  
4 CMC construction and the PMF-MLR model in conjunction with the cluster analysis of back  
5 trajectory. All variables of PM<sub>2.5</sub> mass, their chemical compositions identified, as well as the  
6 sources predicted, were further analysed using correlation matrices with the meteorological-  
7 gaseous pollutants for comprehensive assessment.

8

## 9 **2 Material and methods**

### 10 **2.1 Sampling site description**

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

24 Overall, Peninsular Malaysia experiences relatively uniform temperature (~28.5 °C), high  
25 humidity (more than 70%) and copious rainfall (6.27-15.1mm) throughout the year. Wind  
26 flow pattern distinguishes the seasons for Peninsular Malaysia, namely the South-west (SW)  
27 monsoon, the North-east (NE) monsoon and two shorter periods of inter-monsoons (INT.2  
28 and INT.1) (METMalaysia, 2013). During the SW monsoon (usually established during the  
29 middle of May until the middle of September), the prevailing wind flow is generally south-  
30 westerly and light (below 7.72 m s<sup>-1</sup>). Known as the dry season, haze is expected to occur

1 during this period. On the other hand, during the NE monsoon (established early November  
2 until the middle of March), steady easterly or north-easterly winds of 5.14 to 10.3 m s<sup>-1</sup>  
3 prevail. During periods of strong surges of cold air from the north (cold surges), the winds  
4 over the east coast states of Peninsular Malaysia may reach 15.4 m s<sup>-1</sup> or more. With the  
5 highest rainfall intensity and the possibility of flooding, NE monsoon is known as the wet  
6 season. In this study, air pollution episodes are defined considering PM<sub>2.5</sub> mass (more than 40  
7 µg m<sup>-3</sup>) and the Air Pollution Index (API) (more than 50), hereafter defined as HAZE  
8 samples. Local wind rose, seasonal regional synoptic wind field and biomass fire hotspots are  
9 given in Fig. S1. The average temperature (T) at the site during the sampling campaign was  
10 28.5 ± 1.19 °C and the average relative humidity (RH) was 71.2 ± 7.91%. Following the trend  
11 of T and API, WS was highest during the SW monsoon at an average of 1.39 ± 0.187 m s<sup>-1</sup>  
12 and lowest during the NE monsoon at 1.20 ± 0.167 m s<sup>-1</sup> with an annual average of 1.29 ±  
13 0.194 m s<sup>-1</sup>. Rainfall was lowest during the SW monsoon (6.27 ± 10.6 mm) and highest during  
14 the NE monsoon (15.1 ± 22.7 mm). Overall, the main wind direction for the site was south-  
15 easterly, that is East-South-East (ESE), South-East (SE) and South-South-East (SSE). Details  
16 of the meteorological and gaseous pollutants for each season are given in Table S1.

## 17 **2.2 Aerosol sampling**

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

1 including 12 fields blank (one for each month). The samples were stored at  $-18\text{ }^{\circ}\text{C}$  in a  
2 freezer prior to analysis.

### 3 **2.3 Chemical analyses**

#### 4 **2.3.1 Major ions**

5 For the purpose of soluble ion analysis, one strip ( $2.54\text{ cm} \times 20.32\text{ cm}$ ) of loaded quartz filter  
6 was used. The portion was cut into smaller pieces ( $1\text{ cm} \times 1\text{ cm}$ ) directly into a 50 ml conical  
7 flask. 20 ml of ultra-pure water, UPW (Hach, Millipore Direct-Q 3 UV System; USA) with a  
8 resistivity of  $18.2\text{ M}\Omega$  were added and the flask capped with a stopper. For sonication  
9 extraction purposes ( $60\text{ }^{\circ}\text{C}$ ; 60 m), an ultrasonic bath (Elma Schmidbauer GmbH; Elmasonic  
10 S40; Germany) was used. The solution was subsequently filtered through  $0.2\text{ }\mu\text{m}$  25 mm  
11 Acrodisc filters (Pall; Part number 4612; USA) using a 20 cc/ml Terumo syringe directly into  
12 a 25 ml volumetric flask, class A. UPW was added to the solution to the mark. The solutions  
13 were then directly transferred into two sets of 12 ml centrifuge tubes for separate anion and  
14 cation analysis. The extracted solutions were stored overnight in a refrigerator at  $4\text{ }^{\circ}\text{C}$  to allow  
15 for equilibrium of the solution before analysis using ion chromatography (IC). The analysis  
16 took place within 48 h of extraction. Anion ( $\text{F}^{-}$ ,  $\text{Cl}^{-}$ ,  $\text{NO}_2^{-}$ ,  $\text{Br}^{-}$ ,  $\text{NO}_3^{-}$ ,  $\text{PO}_4^{3-}$ ,  $\text{SO}_4^{2-}$ ) were  
17 analysed using a Metrohm 882 Compact IC plus 1 equipped with column type Metrosep A  
18 Supp 5 – 150/4.0 (Metrohm; USA) while a Metrohm 733 IC Separation Centre (Metrohm;  
19 USA) was used for cation analysis ( $\text{Na}^{+}$ ,  $\text{NH}_4^{+}$ ,  $\text{K}^{+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ). A six-point calibration (0.5,  
20 1, 2, 5, 10 and 20 ppm) was used. The method detection limits (MDL) were calculated based  
21 on three times the standard deviation of field blank ( $n=6$ ) while 1 ppm standard of Single  
22 Cation/Anion Standards (Certipur® Reference Materials for Ion Chromatography, Merck  
23 Millipore, Merck KGaA, Darmstadt, Germany) was used for the calculation of percentage  
24 recoveries. The percentage recoveries for all elements were between 86 and 131%, as reported  
25 in Table S2.

#### 26 **2.3.2 Trace elements**

27 For trace elements, microwave-assisted extraction using acid digestion (4:1 of  $\text{HNO}_3$  and  
28  $\text{H}_2\text{O}_2$ ) was performed using a Milestone Microwave Laboratory System (Gemini BV; MLS-  
29 1200 Mega; Netherlands). For the digestion process, one strip ( $2.54\text{ cm} \times 20.32\text{ cm}$ ) of loaded  
30 filter was used with the following setting of time (m) and power (W) was used: 1) 1, 250; 2)



1 1, 0; 3) 8, 250; 4) 5, 400 and 5) 5, 650. The solution was subsequently filtered through 0.2  $\mu\text{m}$   
2 25 mm Acrodisc filters (Pall Gelmann) using a 50cc  $\text{ml}^{-1}$  Terumo syringe directly into a 50 ml  
3 Teflon volumetric flask. This solution was then topped up with UPW to the mark before  
4 transfer into a 60 ml high density polyethylene (HDPE) bottle for storage. These stocks were  
5 kept in a refrigerator at 4  $^{\circ}\text{C}$  before analysis. Analysis of the elements was carried out using  
6 inductively coupled plasma mass spectrometry, ICPMS (PerkinElmer Instrument; Model Elan  
7 9000; USA). MDL was estimated as three times the standard deviation of field blank ( $n=6$ )  
8 while 1 ppm Multi-Element Calibration Standard 3 (PerkinElmer Pure Plus, PerkinElmer;  
9 USA) was use for validation purpose. Percentage recoveries are based on SRM1648a Urban  
10 PM (National Institute of Standards and Technology, MD, USA) and these varied between 29  
11 and 101%, as reported in Table S2. Details of experimental quality assurance and quality  
12 control (QA/QC) for both trace elements and major ions are provided in the Supplement.

### 13 **2.3.3 Black carbon**

14 BC concentration was determined using a Smokestain Reflectometer with calibration  
15 (Diffusion Systems Ltd.; Model EEL 43M; United Kingdom). In brief, this method involves  
16 the measurement of the darkness of the stain (on the filter paper) through its reflectance of  
17 white light (using a reflectometer). The reflectance is relative to the light reflected by a clean  
18 filter of the same material with an assumption of 100% reflection. The absorbed light then  
19 converted (through calculation) for BC mass. In this study, five points throughout the filters  
20 were taken where the average was then used as the final measured percentage of reflectance  
21 for mass calculation. Additional explanations pertaining to this instrument and the calculation  
22 involved have been discussed elsewhere (Wiwolwattanapun et al., 2011; Moldanová et al.,  
23 2013).

24

### 25 **2.4 Meteorological-gaseous measurements**

26 All meteorological parameters and gaseous pollutants were obtained from the Air Quality  
27 Division of the DOE, Ministry of Natural Resources and Environment of Malaysia. The  
28 meteorological parameters included temperature (T), RH, wind speed (WS), WD and daily  
29 values of API readings while the gaseous pollutants were carbon monoxide (CO), ozone ( $\text{O}_3$ ),  
30 sulfur dioxide ( $\text{SO}_2$ ), nitrogen oxides ( $\text{NO}_x$ ), nitrogen monoxide (NO) and nitrogen dioxide

1 (NO<sub>2</sub>). The instrument and measurement principle used for the gaseous were as follows: O<sub>3</sub> =  
2 Analyzer 400A (chemiluminescence); NO, NO<sub>2</sub>, NO<sub>x</sub> = Teledyne Advanced Pollution  
3 Instrumentation 200A (chemiluminescence); SO<sub>2</sub> = Teledyne Advanced Pollution  
4 Instrumentation M100A (fluorescence); and CO = Teledyne Advanced Pollution  
5 Instrumentation M300 (non-dispersive infrared absorption). API for Malaysia is calculated  
6 based on five major air pollutants including SO<sub>2</sub>, NO<sub>2</sub>, CO, PM<sub>10</sub> and O<sub>3</sub>. These  
7 measurements were recorded at a station (registered station for the DOE Malaysia) less than 1  
8 km south from our sampling location. Details of the monitoring equipment and procedures  
9 involved have been described by (Khan et al., 2015a). Daily rainfall readings, daily PM<sub>10</sub>  
10 (particles with aerodynamic diameter less than 10 μm) and TSP (total suspended particulate)  
11 mass (high volume sampler) were obtained from MET of Petaling Jaya recorded at the  
12 sampling site.

13

## 14 **2.5 Data analysis and modelling**

### 15 **2.5.1 Statistical and diagram plot**

16 All descriptive and statistical analyses were carried out using either PASW Statistics for  
17 Windows, Version 18 (SPSS, 2009) or using Microsoft® Excel 2010 (Excel, 2010) with the  
18 statistical add-in XLSTAT Version 2014.3.04 (Addinsoft, 2014). Meteorological analysis for  
19 monsoonal effects was conducted with the application of several adapted analysis software  
20 packages. For wind vectors, the Grid Analysis and Display System (GrADS version 2.0.2)  
21 was used. The synoptic wind fields were plotted using a dataset (u, v - wind) downloaded  
22 from the National Center for Environmental Protection (NCEP) / National Center for  
23 Atmospheric Research (NCAR)  
24 (<http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.pressure.html>). The dataset  
25 downloaded was selected at 925 hPa (500 m) with a mapping covering latitude: -10°, 20° N,  
26 longitude: 90°, 120° E. For biomass hotspots, fire data from the Moderate-resolution Imaging  
27 Spectroradiometer (MODIS) representing the biomass burning hotspots in the specific area of  
28 interest was used. Data were downloaded from the National Aeronautics and Space  
29 Administration-Land Atmosphere Near Real-time capability for Earth Observing System  
30 (EOS)-Fire Information for Resource Management System (NASA LANCE FIRMS) fire

1 archive (<https://firms.modaps.eosdis.nasa.gov/download/request.php>) in the range of 10 °S to  
2 20 °N and 90 °W to 120 °E. These data were then appended on the map plotted using Igor Pro  
3 6.22A (WaveMetrics, USA). In addition, 48 h backward trajectories were also included onto  
4 the same map using the Hybrid Single-Particle Lagrangian Integrated Trajectory Model  
5 (HYSPLIT 4.9). To ensure consistency with the wind field, the trajectory release was chosen  
6 at about 925 hPa (500 m) with 6 h trajectory intervals were selected. For local wind roses (for  
7 each season), which were plotted using Igor Pro 6.22A (WaveMetrics, USA), data obtained  
8 from the DOE were used.

## 9 **2.5.2 Chemical mass closure**

10 Modified from Bressi et al. (2013), seven major groups were considered for the CMC  
11 calculations: sea salt (ss), dust, secondary inorganic aerosol (SIA), trace element (TE), BC, K<sup>+</sup>  
12 and also the unidentified portion of the PM mass. Due to our low Al element recovery (36%),  
13 and lack of Si and S elements which are the dominant elements in soil from PM<sub>2.5</sub> (Rahman et  
14 al., 2011) the dust fraction is therefore calculated using a straightforward approach used by  
15 Bressi et al. (2013). The dust fraction was calculated as the contribution of nss-Ca<sup>2+</sup> in mineral  
16 dust. The 8.3% mineral dust mass contribution for the Klang Valley area estimated by  
17 Rahman et al. (2011) was employed for the calculation. Following the direct CMC nss-Ca<sup>2+</sup>  
18 approach, we therefore exclude the major mineral dust elements (Al, Fe) to calculate the rest  
19 of trace element mass contribution.

20 The overall calculations involved for the CMC were as follows:

$$21 \text{ [PM}_{2.5}\text{]} = \text{[Sea salt]} + \text{[Dust]} + \text{[SIA]} + \text{[TE]} + \text{[BC]} + \text{[K}^+\text{]} + \text{[Unidentified]} \quad (1)$$

22 where,

$$23 \text{ [Sea salt]} = \text{[Na}^+\text{]} + \text{[Cl}^-\text{]} + \text{[Mg}^{2+\text{]} + \text{[ss-K}^+\text{]} + \text{[ss-Ca}^{2+\text{]} + \text{[ss-SO}_4^{2-\text{]};$$

$$24 \text{ with [ss-K}^+\text{]} = 0.036 \times \text{[Na}^+\text{]}; \text{ [ss-Ca}^{2+\text{]} = 0.038 \times \text{[Na}^+\text{]}; \text{ and}$$

$$25 \text{ [ss-SO}_4^{2-\text{]} = 0.252 \times \text{[Na}^+\text{]}$$

$$26 \text{ [Dust]} = \text{[nss-Ca}^{2+\text{]} / 0.083$$

$$27 \text{ [SIA]} = \text{[nss-SO}_4^{2-\text{]} + \text{[NO}_3^-\text{]} + \text{[NH}_4^+\text{]};$$

$$28 \text{ with [nss-SO}_4^{2-\text{]} = \text{[SO}_4^{2-\text{]} - \text{[ss-SO}_4^{2-\text{]}; \text{ “nss-” standing for “non-sea salt”}$$

### 1 **2.5.3 Enrichment factor**

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

### 11 **2.5.4 Source apportionment**

12 A combination of PMF version 5.0 (PMF 5.0) and multilinear regression (MLR) analysis was  
13 employed to determine source apportionment where results of the MLR were used to  
14 apportion the PM<sub>2.5</sub> chemical compositions in order to quantify sources. Details of the PMF  
15 procedure used in this study are similar to our previous work as discussed in Khan et al.  
16 (2015b). In brief, two data files were used as an input, i.e. 1) concentration; and 2)  
17 uncertainty. For the concentration data file, the chemical composition dataset were first pre-  
18 treated and validated. To ensure a strong signal from the data was evident, species having  
19 more than 50% of the data below MDL were discarded. For the rest, the missing values were  
20 replaced by half of the MDL while data with values, but below MDL, were left as they were.  
21 The final dataset used for the PMF analysis contained 80 samples with 31 elements (including  
22 PM<sub>2.5</sub> mass) whereby only elements that have high recovery were used. Based on the signal-  
23 to-noise (S/N) ratio, NO<sub>3</sub><sup>-</sup> and Na<sup>+</sup> were set as ‘weak’ species ( $0.2 \leq S/N < 2$ ) while the rest  
24 were categorised as ‘strong’ species (ratio  $\geq 2$ ) (Heo et al., 2009; Richard et al., 2011; Yu et  
25 al., 2013). The PM<sub>2.5</sub> mass was also categorised as “weak” so as not to affect the PMF  
26 solution. The second data file is the uncertainty value of each variable in each sample  
27 estimated from an empirical equation. An additional 5% uncertainty was added to account for  
28 methodological errors during preparation of filter papers, gravimetric mass measurements and  
29 preparing the calibration curves. Upon running the PMF analysis, different numbers of factors  
30 and Fpeak values have been explored to obtain the most meaningful results with 100 bootstrap  
31 runs and a minimum R<sup>2</sup> of 0.6 to test the uncertainty of the resolved profiles. Fpeak is a

1 parameter uses to execute the test of rotational ambiguity and the bootstrap is used to detect  
2 and estimate the disproportionate effects or errors of a small set of observation resampling  
3 randomly. Paatero et al. (2014) suggested that each resampled version of observation, some  
4 randomly chosen rows of the original matrix occur multiple times, while other rows do not  
5 occur at all. Similar to base run, each resampled data set is decomposed into profile and  
6 contribution matrices using PMF. Our observations of each PMF run using the each starting  
7 point of seed from 1 until 9 are summarized in Table S3. The results showed that the solution  
8 was rather stable at the starting point of seed 9. It was observed that a 5 factor solution  
9 provided the most meaningful results, based on the lowest Q (Robust) and Q (True) value of  
10 1581.27 with the Q (true)/Q exp value of 0.94 after 390 computational steps and the  
11 convergence of the results. PMF factors were resolved on 20 runs and seed value of 9, with  
12  $F_{\text{peak}} = 0$  found to be the most reasonable. The model output of source contribution is  
13 provided as normalised or dimensionless (average of each factor contribution is one). To  
14 express the output of PMF, the mass concentrations of the identified sources were scaled by  
15 using the MLR analysis.

16

### 17 **3 Results and discussion**

#### 18 **3.1 PM<sub>2.5</sub> mass and its relations to meteorological and gaseous conditions**

##### 19 **3.1.1 PM<sub>2.5</sub> mass variations**

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

1 with  $17 \mu\text{g m}^{-3}$  during the NE monsoon. Among the weekdays, Friday recorded the highest  
2 average value of  $\text{PM}_{2.5}$  mass at  $33 \mu\text{g m}^{-3}$  while lowest was on Wednesday ( $24 \mu\text{g m}^{-3}$ ).  
3 Meanwhile, weekends on average recorded lower  $\text{PM}_{2.5}$  mass ( $26 \mu\text{g m}^{-3}$ ) compared to  
4 weekdays ( $29 \mu\text{g m}^{-3}$ ).

5  $\text{PM}_{2.5}$  mass shows significant variability between the NE monsoon and the three other seasons  
6 (SW, INT.2 and INT.1). Figure 2c showed that during the NE monsoon, only 17%  
7 exceedance of the daily WHO guideline was recorded while for three other seasons, more than  
8 50% exceedance of the daily WHO guideline was recorded. The small number of exceedances  
9 during the NE monsoon was due to high rainfall (precipitation) during this time. Juneng et al.  
10 (2009) and Rashid and Griffiths (1995) also reported similar observations of seasonal  
11 fluctuation of particulate concentration with minimal concentration during the rainy season of  
12 the NE monsoon. Most exceedance days occurred during the dry seasons of the SW monsoon  
13 and INT.2 (middle May until end of October) with 66% and 71% exceedance, respectively.  
14 Similar observations of high exceedances during the SW monsoon dry season have been  
15 recorded for Peninsular Malaysia in general and the Klang Valley in particular (Rashid and  
16 Griffiths, 1995; Juneng et al., 2011; Norela et al., 2013; Tahir et al., 2013b; Amil et al.,  
17 2014). Higher mass concentrations during the dry season were also seen in other SEA (Kim  
18 Oanh et al., 2006; Lestari and Mauliadi, 2009) and Asian cities (Reid et al., 2013). As shown  
19 in Fig. 2a, it is important to note that haze events always occur during the SW monsoon, thus  
20 it is anticipated that they will directly affect the SW overall mass concentration ( $\text{PM}_{2.5}$ ).  
21 However, the ANOVA analysis showed that HAZE is significantly different from the SW  
22 monsoon on an overall perspective ( $p = 0.003$ ). This is perhaps due to short pollution episodes  
23 (HAZE) compared to the long period of the SW monsoon. HAZE events for this study  
24 averaged at  $61 \pm 24 \mu\text{g m}^{-3}$ , higher compared to the 2011 haze episode documented for Bangi  
25 area at  $48 \pm 10 \mu\text{g m}^{-3}$  by Amil et al. (2014).

26 The annual  $\text{PM}_{2.5}$  mass (weekly average representative of the month) for this study averaged  
27 at  $28 \pm 18 \mu\text{g m}^{-3}$ . This is almost triple (2.8 fold) the  $10 \mu\text{g m}^{-3}$  WHO  $\text{PM}_{2.5}$  annual guideline,  
28 2.33 fold higher than the US EPA NAAQS  $\text{PM}_{2.5}$  annual standard of  $12 \mu\text{g m}^{-3}$  and 1.12 fold  
29 higher than the European Union (EU)  $\text{PM}_{2.5}$  annual standards set at  $25 \mu\text{g m}^{-3}$  (European  
30 Commission, 2015). Table 2 reports that  $\text{PM}_{2.5}$  mass average for this study was very low  
31 compared to other big cities of Asia i.e. in India and China (Balakrishnaiah et al., 2012;  
32 Huang et al., 2013; Pachauri et al., 2013; Zhang et al., 2013) but variable when compared to

1 other parts of the world (Dongarrà et al., 2010; Yin et al., 2010; Bressi et al., 2013;  
2 Squizzato et al., 2013). On a local scale, the average value of PM<sub>2.5</sub> mass for the site was  
3 slightly higher than previous measurements carried out here during 2004 – 2008 ( $27 \pm 10 \mu\text{g}$   
4  $\text{m}^{-3}$ ) (Rahman et al., 2011) but lower compared to measurements carried out during 1998 –  
5 2000 ( $33 \mu\text{g m}^{-3}$ ) (Keywood et al., 2003). Furthermore, our result for Petaling Jaya is higher  
6 than other parts of Peninsular Malaysia (Tahir et al., 2013b; Ee-Ling et al., 2015).

7 The mean PM<sub>2.5</sub>/PM<sub>10</sub> ratio for the site was  $0.72 \pm 0.18$  and the ratio for PM<sub>2.5</sub>/TSP was  $0.46 \pm$   
8  $0.13$ , as reported in Table 1. PM<sub>10</sub>/TSP ratio was  $0.63 \pm 0.12$ . The PM<sub>2.5</sub>/PM<sub>10</sub> ratio at this site  
9 was higher than other studies in Asia as reported by Hopke et al. (2008) where most of the  
10 sites studied showed ratios of lower than 0.50. From the aforementioned study, however, an  
11 urban site in China and suburban site in Lembang, Indonesia recorded similar PM<sub>2.5</sub>/PM<sub>10</sub>  
12 ratio to our result of more than 0.70. Our PM<sub>2.5</sub>/PM<sub>10</sub> ratio was also in agreement with other  
13 cities in Europe (Gehrig and Buchmann, 2003; Gomišček et al., 2004; Contini et al., 2014).  
14 Despite having different characteristics, the SW and NE monsoons still came out with similar  
15 values to the annual PM<sub>2.5</sub>/PM<sub>10</sub> ratio at  $0.72 \pm 0.10$  and  $0.71 \pm 0.13$ , respectively. The similar  
16 PM<sub>2.5</sub> to PM<sub>10</sub> ratio during the wet and the dry season indicates that meteorological  
17 parameters, specifically rainfall, are affecting the fine (particle with an aerodynamic diameter  
18 of less than 2.5  $\mu\text{m}$ ) and coarse (particle with an aerodynamic diameter of greater than 2.5  
19  $\mu\text{m}$ ) particles in the same way. This is also confirmed by the good correlation of PM<sub>2.5</sub> and  
20 PM<sub>10</sub> ( $r = 0.963$ ;  $p < 0.0001$ ). Both inter-monsoon seasons recorded the opposite mass  
21 concentration trend. INT.2 (average mass of  $29 \pm 12 \mu\text{g m}^{-3}$ ) showed a higher mass  
22 concentration than INT.1 (average mass of  $23 \pm 8 \mu\text{g m}^{-3}$ ) but a lower PM<sub>2.5</sub>/PM<sub>10</sub> ratio ( $0.62$   
23  $\pm 0.17$ ) than INT.1 ( $0.85 \pm 0.40$ ). This ratio of INT.1 is the highest PM<sub>2.5</sub>/PM<sub>10</sub> ratio among all  
24 seasons, even higher than during HAZE episodes. HAZE-episode-only ratios were  $0.74 \pm$   
25  $0.070$ . To further examine the particle at the site, the seasonal PM<sub>2.5</sub>/TSP ratio was calculated.  
26 During the dry season (the SW monsoon), ambient air at the site had particles in the ratio of  
27 approximately 50/50 coarse to fine particles (PM<sub>2.5</sub>/TSP =  $0.50 \pm 0.081$ ). During INT.2 and  
28 the NE monsoon (wet season), the air was filled with more coarse particles, resulting in  
29 PM<sub>2.5</sub>/TSP ratios of  $0.44 \pm 0.12$  and  $0.40 \pm 0.087$ , respectively. INT.1 and HAZE episodes on  
30 other hand both had a PM<sub>2.5</sub>/TSP ratio of 0.54, implying the ambient air contained almost the  
31 same portion of fine and coarse particles. With these ratios, we can conclude that fine particles  
32 are very significant in the ambient air of the Petaling Jaya urban-industrial area in Klang

1 Valley. Similar observation on the significance of the fine particle were also reported for SEA  
2 cities (Kim Oanh et al., 2006).

### 3 **3.1.2 Relationship between PM<sub>2.5</sub> and meteorological-gaseous influence**

4 Referring to Table 3, the Pearson correlation revealed that PM<sub>2.5</sub> mass on an annual basis was  
5 significantly influenced by meteorological and gaseous parameters. Among the parameters,  
6 API strongly correlated with PM<sub>2.5</sub> mass ( $r = 0.763$ ;  $p < 0.001$ ). Since the Malaysian API  
7 includes PM<sub>10</sub>, this result was anticipated due to the high ratio of PM<sub>2.5</sub>/PM<sub>10</sub> (0.72). The  
8 PM<sub>2.5</sub> mass was positively correlated with T ( $r = 0.310$ ;  $p = 0.005$ ) and negatively correlated  
9 with RH ( $r = -0.314$ ;  $p < 0.005$ ). Having used wind flow to distinguish the season for  
10 Malaysia, the WS influence towards the PM<sub>2.5</sub> mass was as expected ( $r = 0.274$ ;  $p < 0.05$ ).  
11 However, rainfall and WD did not significantly correlate with PM<sub>2.5</sub> mass at the site. With an  
12 exception of NO<sub>x</sub>, all other gaseous parameters were found to significantly influence the  
13 PM<sub>2.5</sub> mass. CO and NO<sub>2</sub> were significantly positively correlated with PM<sub>2.5</sub> ( $p < 0.0001$ ) at  $r$   
14 = 0.471 and  $r = 0.473$  respectively, indicating a combustion-related traffic source. The  
15 significant positive correlation between PM<sub>2.5</sub> and SO<sub>2</sub> ( $r = 0.324$ ;  $p < 0.005$ ) further supports  
16 this. NO was the only gaseous parameter that had a negative relationship with PM<sub>2.5</sub> mass ( $r =$   
17  $-0.262$ ;  $p < 0.0001$ ). O<sub>3</sub> on the other hand showed a significant positive correlation with PM<sub>2.5</sub>  
18 mass at  $r = 0.298$  ( $p < 0.01$ ). The significant positive correlation of PM<sub>2.5</sub> and O<sub>3</sub> possibly  
19 indicates a secondary source of PM<sub>2.5</sub> as well as the already identified combustion-related  
20 traffic source, which is primary.

21 On a seasonal scale, daily PM<sub>2.5</sub> mass during all seasons appeared to be affected by the  
22 gaseous parameters but not meteorological conditions. PM<sub>2.5</sub> mass during the SW monsoon,  
23 which is also known as the dry season, was strongly correlated with CO ( $r = 0.687$ ;  $p <$   
24  $0.001$ ), O<sub>3</sub> ( $r = 0.535$ ;  $p < 0.005$ ), NO<sub>2</sub> ( $r = 0.528$ ;  $p < 0.05$ ) and API ( $r = 0.748$ ;  $p < 0.001$ ).  
25 NE (the wet season) showed strong correlations with SO<sub>2</sub> and NO<sub>2</sub> with  $r = 0.654$  ( $p < 0.001$ )  
26 and  $r = 0.711$  ( $p < 0.001$ ), respectively. NO showed the least effect towards PM<sub>2.5</sub> mass. Both  
27 INT.2 and INT.1 correlated strongly with NO<sub>2</sub>,  $r = 0.851$  ( $p < 0.001$ ) and  $r = 0.874$  ( $p <$   
28  $0.001$ ), respectively. In addition, INT.2 also showed a significant correlation with NO<sub>x</sub> ( $r =$   
29  $0.800$ ;  $p < 0.001$ ) while INT.1 correlated strongly with CO ( $r = 0.654$ ;  $p < 0.05$ ) and API ( $r =$   
30  $0.705$ ;  $p < 0.05$ ). HAZE episodes, as expected, were significantly correlated with CO ( $r =$   
31  $0.749$ ;  $p < 0.05$ ), which is one of the key pollution tracers. With Malaysia having relatively



1 uniform temperature, high humidity and copious rainfall throughout the year, minimal  
2 influence of meteorological parameters towards seasonal PM<sub>2.5</sub> mass variation is predicted.  
3 Rainfall showed no significant correlation with PM<sub>2.5</sub> mass even during the two seasons of the  
4 SW monsoon (dry season with low RH and rainfall, high WS) and the NE monsoon (wet  
5 season with high RH and rainfall, low WS). However, INT.2 showed a strong negative  
6 correlation with rainfall ( $r = -0.733$ ,  $p > 0.05$ ). This may be due to the transition period of the  
7 WD in between the two monsoons. For the PM<sub>2.5</sub>-T relationship, all four seasons of  
8 Peninsular Malaysia shows positive correlations. HAZE events revealed a slight negative  
9 correlation between PM<sub>2.5</sub> mass and T. This condition is perhaps because during haze  
10 episodes, the small particles envelope the atmosphere and reduce the UV radiation which can  
11 reduce the temperature of earth surface. RH and PM<sub>2.5</sub> mass on the other hand, revealed  
12 negative relationships with three seasons (except INT.1) having low correlations. INT.1  
13 showed the reverse relationship. However, HAZE events which occur during the SW  
14 monsoon, disagree with the generic pattern of the SW monsoon PM<sub>2.5</sub>-RH relationship. WS  
15 and WD on a seasonal scale showed no significant correlation towards PM<sub>2.5</sub> in all four  
16 seasons, even during the HAZE events. As mentioned earlier, Table 1 reported that the  
17 PM<sub>2.5</sub>/PM<sub>10</sub> ratio for both major seasons (SW and NE) were almost the same at ~0.70. The  
18 PM<sub>2.5</sub>/TSP and PM<sub>10</sub>/TSP ratios were different, however. During the SW monsoon ratios of  
19 0.50 and 0.70 were observed, while during the NE monsoon ratios of 0.40 and 0.57 were  
20 recorded for PM<sub>2.5</sub>/TSP and PM<sub>10</sub>/TSP respectively. These ratios support the findings of  
21 meteorological parameters (rainfall, WS and WD) not significantly correlating with PM<sub>2.5</sub>  
22 mass variability with changing season at the site. Instead, results reveal that perhaps  
23 meteorological parameters only greatly influence the coarse particles (PM dp > 2.5 μm) but  
24 not fine particles at the site.

### 25 **3.2 Chemical composition**

26 Referring to Fig. 3a and Table S2, chemical compositions of PM<sub>2.5</sub> determined were water-  
27 soluble ions (anions and cations), trace elements (including heavy metals) and BC for a total  
28 of 36% of PM<sub>2.5</sub> mass. BC accounted for about 15% (4.15 μg m<sup>-3</sup>) of the PM<sub>2.5</sub> mass. The total  
29 anion mass measured was 1.67 μg m<sup>-3</sup> (6.0% of PM<sub>2.5</sub> mass) while the total cation mass was  
30 1.75 μg m<sup>-3</sup> (6.3% of PM<sub>2.5</sub> mass). As shown in Fig. S2, the equivalent charge ratio of total  
31 cation to total anion ratio was 0.46 indicates that the aerosol at the site is acidic due to the

1 excess of anions, also experienced by other study (He et al., 2012). The trend for anions was:  
2  $\text{SO}_4^{2-} > \text{NO}_3^- > \text{PO}_4^{3-} > \text{Cl}^- > \text{Br}^- > \text{NO}_2^- > \text{F}^-$  while the cation trend was:  $\text{NH}_4^+ > \text{Na}^+ > \text{K}^+ >$   
3  $\text{Ca}^{2+} > \text{Mg}^{2+}$ . The overall water-soluble trend for this urban-industrial site was:  $\text{SO}_4^{2-}$  (39% of  
4 water-soluble ions; 23% of IM mass)  $> \text{NH}_4^+$  (29% of water-soluble ions; 17% of IM mass)  $>$   
5  $\text{Na}^+$  (9% of water-soluble ions, 5% of IM mass)  $> \text{K}^+$  (7% of water-soluble ions; 4% of IM  
6 mass)  $> \text{NO}_3^-$  (6% of water-soluble ion; 4% of IM mass)  $> \text{Ca}^{2+} > \text{PO}_4^{3-} > \text{Mg}^{2+} > \text{Cl}^- > \text{Br}^- >$   
7  $\text{NO}_2^- > \text{F}^-$ . Trace elements on the other hand accounted for about 8.6% of  $\text{PM}_{2.5}$  mass ( $2.41 \mu\text{g}$   
8  $\text{m}^{-3}$ ) with the major elements Al (44% of TE), Fe (42%), Zn (8%), and Pb (4%). The rest of  
9 the trace elements were in the decreasing order of: Ba  $> \text{Cr} > \text{Cu} > \text{Rb} > \text{Mn} > \text{V} > \text{Ni} > \text{As} >$   
10  $\text{Sr} > \text{Ag} > \text{Cd} > \text{Se} > \text{Ga} > \text{Cs} > \text{Bi} > \text{Co} > \text{Li} > \text{U} > \text{Be}$ . It is notable that results for Pb, As, Cd  
11 and Ni in this study did not exceed any EU standard on air pollutants. The 8.6% mass  
12 percentage of trace elements determined in this Petaling Jaya urban-industrial site is lower  
13 than the 14.% trace element recorded at Kuala Lumpur city (Rahman et al., 2011) but higher  
14 compared to Kuala Terengganu (Tahir et al., 2013b).

15

## 16 **Chemical mass closure**

17 For a better understanding of the  $\text{PM}_{2.5}$  chemical variability on a seasonal scale, we  
18 constructed a CMC on proportions of all identified components; as illustrated in Fig. 3b. In  
19 general, the inorganic seasonal variability in  $\text{PM}_{2.5}$  composition is relatively small with both  
20 primary and secondary components of  $\text{PM}_{2.5}$  equally important. In this study, IM accounted  
21 for 19% of  $\text{PM}_{2.5}$  mass while BC accounted for 15%. Therefore, 66% was left unidentified  
22 which was presumed to be sulfur compounds (S) and organic matter. The components for the  
23 aforementioned inorganic portion were as follows: SIA ( $2.49 \mu\text{g m}^{-3}$ ; 9%)  $> \text{dust}$  ( $2.09 \mu\text{g m}^{-3}$ ;  
24 7%)  $> \text{TE}$  ( $0.344 \mu\text{g m}^{-3}$ ; 1%)  $> \text{sea salt}$  ( $0.265 \mu\text{g m}^{-3}$ ; 1%)  $> \text{K}^+$  ( $0.253 \mu\text{g m}^{-3}$ ; 1%).

25 SIA, a combination of nss-sulfate ( $\text{nss-SO}_4^{2-}$ ), ammonium ( $\text{NH}_4^+$ ) and nitrate ( $\text{NO}_3^-$ ), in  $\text{PM}_{2.5}$   
26 maintained a similar portion throughout the year – between 8 to 10%, with the highest portion  
27 during INT.2 and lowest during the HAZE. On an annual as well as a seasonal scale  
28 (including HAZE),  $\text{nss-SO}_4^{2-}$  (annual average =  $1.29 \mu\text{g m}^{-3}$ ; 5% of  $\text{PM}_{2.5}$  mass; 23% of IM  
29 mass) was the major SIA component followed by  $\text{NH}_4^+$  (annual average =  $0.987 \mu\text{g m}^{-3}$ ; 4%  
30 of  $\text{PM}_{2.5}$  mass; 17% of IM mass) and  $\text{NO}_3^-$  ( $0.213 \mu\text{g m}^{-3}$ ; 1% of  $\text{PM}_{2.5}$  mass; 4% of IM mass).  
31 Total SIA on this site was 73% of the total water-soluble ions measured, which is lower

1 compared to 79% in Greece (Remoundaki et al., 2013) and 85% in Italy (Squizzato et al.,  
2 2013). The value of nss-SO<sub>4</sub><sup>2-</sup> (97% of SO<sub>4</sub><sup>2-</sup>) and nss-K<sup>+</sup> (96% of K<sup>+</sup>) in this study are almost  
3 the same as results from 2004-2008 by Keywood et al. (2003) at 98% for both nss-SO<sub>4</sub><sup>2-</sup> and  
4 nss-K<sup>+</sup> which is why SO<sub>4</sub><sup>2-</sup> and K<sup>+</sup> were used for PMF SA instead of nss-SO<sub>4</sub><sup>2-</sup> and nss-K<sup>+</sup>.  
5 These results, however, are different from another local study (Tahir et al., 2013a) where nss-  
6 SO<sub>4</sub><sup>2-</sup> and nss-K<sup>+</sup> at a coastal area only made up about 53% and 13% respectively. Hence, we  
7 could draw a conclusion that the SIA at the site is influenced by anthropogenic activities  
8 rather than marine sources even though the Malacca Straits are only about 33 km away.  
9 Following the SIA trend, nss-SO<sub>4</sub><sup>2-</sup> was highest (6%) during INT.2 which is the start of the  
10 rainy season. Surprisingly, the SW and NE monsoons came out with the same nss-SO<sub>4</sub><sup>2-</sup>  
11 portion in PM<sub>2.5</sub> (5%) even though the two have significant differences in terms of  
12 meteorological conditions, especially WD and rainfall; refer to Fig. S1a,c for synoptic wind  
13 direction. NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> also do not vary largely with season, portioned at 4% and 1%;  
14 respectively. HAZE recorded the lowest NO<sub>3</sub><sup>-</sup> portion in PM<sub>2.5</sub> at below half a percent while  
15 NH<sub>4</sub><sup>+</sup> was lowest during the NE monsoon. Also known as the acidity ratio, the neutralisation  
16 ratio (NR) was calculated to further investigate the acidity of the atmospheric aerosols, as  
17 reported in Table S1. The NR was calculated based on the ratio of the NH<sub>4</sub><sup>+</sup> (eq m<sup>-3</sup>) to the  
18 sum of SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> (eq m<sup>-3</sup>) (Squizzato et al., 2013). The overall NR obtained for this site  
19 was 0.26, indicating an excess of SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>. The NR ratio varied with season. The  
20 highest recorded NR was during the HAZE episodes with 0.35. The rest of the values showed  
21 the following trend: SW (0.31) > NE (0.22) > INT.2 (0.21) > INT.1 (0.17).

22 Trace elements, which are good indicators for anthropogenic factors, had a mass contribution  
23 of 0.344 µg m<sup>-3</sup> (1%) on an annual basis with the following seasonal trend: INT.2 (2%) > NE  
24 (2%) > INT.1 (1%) > SW (1%) > HAZE (1%). Referring to the EF analysis (Fig. S3), most of  
25 the metals studied can be assumed to originate from anthropogenic sources, i.e. Pb, Se, Zn,  
26 Cd, As, Bi, Ba, Cu, Rb, V and Ni. Other metals, i.e. Sr, Mn, Co, and Li, are considered to  
27 originate from crustal sources. Pb, Zn, Cu, Cd, V and Ni are reflecting the traffic sources. Co,  
28 Sr and Li are typical soil constituents (Pey et al., 2009). Following Kuo et al. (2007), the  
29 elements can be categorised based on the degree of enrichment which in this study the annual  
30 EF gives the following results: 1) highly enriched (EF ≥ 1000): Pb; 2) moderately enriched  
31 (100 < EF < 1000): Se, Zn and Cd; 3) slightly enriched (10 < EF < 100): As, Bi and Ba; and  
32 4) minimally enriched (EF < 10): Cu, V, Ni, Sr, Mn, Co and Li. However, the seasonal results

1 revealed a slight difference in several elements (Cu, Rb, V and Ni); as shown in Fig. S3. For  
2 example, Cu during SW monsoon follows the annual grouping of anthropogenic source while  
3 during other seasons, it is drawn from the crustal source. Meanwhile, Rb, V and Ni during the  
4 SW monsoon originate from the anthropogenic source which is contrary to the annual and  
5 other seasonal patterns. Ni and V are known as heavy oil combustion indicators (Jiang et al.,  
6 2014), Cu is known to be associated with the traffic (Contini et al., 2014) while Rb is known  
7 to be drawn from the crustal source (Khan et al., 2010a). A study in Taiwan also argued that  
8 these four elements (Cu, Rb, V and Ni) are likely to be affected by both soil and non-soil  
9 emissions (Balakrishnaiah et al., 2012).

10 Dust was one of the minor mass components of PM<sub>2.5</sub> and averaged at 7% on an annual basis.  
11 This component showed the highest percentage during INT.2 (9%), decreased a little in the  
12 following NE monsoon (7%), continued to decrease in the INT.1 (6%) and increased back  
13 again during the following SW monsoon (9%). The HAZE episodes, however, recorded the  
14 lowest dust portion in PM<sub>2.5</sub> at 6%. The seasonal patterns of dust portions relate to the  
15 meteorological conditions. During the NE monsoon the wind is blown from the Siberian High  
16 (Siberian Anticyclone) over Southeast Asia i.e. Southern-China (Indo-China), Cambodia,  
17 Vietnam and the Philippines while during the SW monsoon, the wind flow is from Australia  
18 and neighbouring countries, i.e. Singapura and Indonesia (especially Sumatra and Jawa  
19 Island); as shown in Fig. S1a, c.

20 Sea salts form only ~1% of PM<sub>2.5</sub> mass on an annual scale confirming the findings of a  
21 previous study by Keywood et al. (2003). Seasonally, the percentage remains below 1%  
22 except during INT.1 where the sea salt portion is highest (4%). However, the specific  
23 percentage value still shows the difference where the NE, and SW monsoons, INT.2 and  
24 HAZE portion at 0.99, 0.38, 0.28 and 0.18, respectively. The low percentage of sea salt in  
25 PM<sub>2.5</sub> is similar to the findings of a study by Tahir et al. (2013a) which observed that nss-ionic  
26 species accounted for 88% of the total ions associated with PM<sub>2.5</sub>. PM<sub>2.5</sub> at this site is expected  
27 to have a low marine contribution because marine aerosol is typically associated with coarse  
28 particles as seen by Tahir et al. (2013b) and Almeida et al. (2005). Khan et al. (2010b) also  
29 reported similar observations where the four major marine elements, Na<sup>+</sup>, Cl<sup>-</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>,  
30 were dominant in coarse particles (PM<sub>2.5-10</sub> and PM<sub>>10</sub>). K<sup>+</sup>, which is normally recognised as  
31 the biomass burning indicator, represented only 1% of PM<sub>2.5</sub> mass (0.253 ± 0.144 µg m<sup>-3</sup> on  
32 annual scale) regardless of the season change including the HAZE episodes.

1 BC averaged at  $4.15 \pm 0.642 \mu\text{g m}^{-3}$  (15% of  $\text{PM}_{2.5}$  mass). The highest proportion was seen  
2 during the rainy season of the NE monsoon (21%) and lowest during the dry season of the SW  
3 monsoon (11%). The HAZE events showed a result of 8%. The two inter-monsoons seasons  
4 recorded average values between the two major seasons. Also known as elemental carbon  
5 (EC) (Lanz et al., 2010), the BC result measured here is within the range of Malaysia's initial  
6 results on BC measured at the same site by Abas and Simoneit (1996). They found  $9 \mu\text{g m}^{-3}$   
7 EC with  $74 \mu\text{g m}^{-3}$  of organic carbon (OC) in TSP samples (TSP mass of  $300 \mu\text{g m}^{-3}$ ) during  
8 haze episodes; while during normal days they found 8 and  $14 \mu\text{g m}^{-3}$  for EC and OC  
9 respectively from  $74 \mu\text{g m}^{-3}$  of TSP mass. The BC value for this study (annual = 15%, HAZE  
10 = 8%) was low compared to measurements at the same site during a 1998 – 2000 study by  
11 Keywood et al. (2003). However, our results showed a similar pattern where BC during  
12 HAZE events was lower by at least one third (1/3) compared to normal days (normal = 30%,  
13 haze = 20%). The BC portion here was also similar to measurements carried out in 2004 –  
14 2008 by Rahman et al. (2011) at 15.8%. On a regional scale, our results here are low  
15 compared to most other SEA cities as reported by Reid et al. (2013). One possible reason is  
16 because this study was carried out on a long-term basis while the others mostly concentrate on  
17 a particular season and/or event, especially haze episodes.

18 On an annual scale, the unidentified components reached 66% of the total  $\text{PM}_{2.5}$  mass.  
19 Seasonal variability was observed, with the smallest in the NE monsoon (58%) during the  
20 intensified rainfall with low WS while the largest portions were during HAZE (77%), when  
21 rainfall was low with high WS. One reason for such high uncertainties in the CMCs is the lack  
22 of OC composition which is one of the major components in  $\text{PM}_{2.5}$ . Previous studies by Tahir  
23 et al. (2013b) and Cohen et al. (2004) also reported similar large unidentified portions of  
24  $\text{PM}_{2.5}$  which were presumed to be of organic composition. A large amount of OC (58%) in  
25  $\text{PM}_{2.5}$  was also reported in India at Ahmedabad (Rengarajan et al., 2011) and in an urban-  
26 industrial area of Agra (Pachauri et al., 2013). Cheng et al. (2010) reported a very high  
27 carbonaceous portion of  $\text{PM}_{2.5}$  in Hong Kong of ~70% for three roadside monitoring sites and  
28 ~48% at the ambient site. The portion of our IM and BC were also low compared to the  
29 previous study of the site by Keywood et al. (2003) with 28 and 30% (normal days),  
30 respectively. A study by Remoundaki et al. (2013) revealed that sulfates and carbonaceous  
31 material are major fractions of  $\text{PM}_{2.5}$ , with 35% and 30%, respectively. Considering only the  
32 identified composition, water absorption of water-soluble components may lead to a positive

1 bias during weighing, even in a controlled environment (i.e. RH)(Speer et al., 1997). In  
2 addition, Zhang et al. (2013) mentioned that the volatilisation of  $\text{NH}_4\text{NO}_3$  and organic matter  
3 may result in negative biases in the specific components. This is likely to happen during the  
4 major seasons of the NE and SW monsoons.

### 5 **3.3 Source apportionment and its relation to meteorological-gaseous** 6 **conditions**

7 Referring to Table 4 and Fig. 4a, the PMF 5.0 model resolved five factors, identified as: 1)  
8 combustion of engine oil; 2) mineral dust; 3) mixed SIA and biomass burning; 4) mixed traffic  
9 and industrial; and 5) sea salt; The source contribution by each factor was summed up to  
10 estimate the predicted mass of  $\text{PM}_{2.5}$ . A strong and significant correlation ( $R^2 = 0.901$ ) was  
11 observed as shown by a scatter plot, representing a regression of the predicted and measured  
12  $\text{PM}_{2.5}$  for SA analysis; Fig. 4b. Table 4 summarises the SA results of the relative contributions  
13 from each identified source to the  $\text{PM}_{2.5}$  on a seasonal and annual basis. The dominance of  
14 each identified source largely varies with changing seasons, which is roughly consistent with  
15 the CMC, EF and stoichiometric analysis for a number of factors. Each of the factors is  
16 characterised by a chemical ‘fingerprint’ which is a unique pattern of chemical species and  
17 their concentrations. In addition, we also describe the interpretation SA identified in time  
18 series analysis and its relation to meteorological and gaseous factors (Fig. 5).

#### 19 **3.3.1 Factor 1: combustion of engine oil (V, Sr, Ni, $\text{SO}_4^{2-}$ , Ga, $\text{NH}_4^+$ )**

20 With an annual V/Ni ratio of 1.91, both elements indicate a major contribution of fuel oil  
21 combustion, identified in this study as factor 1. Vanadium in this factor accounts for 53% of  
22 total V mass while Ni represents 51% (of total Ni mass). Many studies have used both  
23 elements as combustion fuel oil indicators (Kowalczyk et al., 1982; Harrison et al., 1996; Ho  
24 et al., 2006; Pey et al., 2009; Jiang et al., 2014). Mueller et al. (2011) indicated that V and Ni  
25 were promising markers for ship engine exhaust while Gibson et al. (2013) identified a  
26 shipping emissions factor based on V, Ni and  $\text{SO}_4^{2-}$  following a study by Zhao et al. (2013).  
27 Since Port Klang (one of the major ports in Malaysia) is about 33 km from our sampling site,  
28 there is a possibility of ship emissions to contributing to this factor. However, a number of  
29 studies have recognised a combination of V, Ni and  $\text{SO}_4^{2-}$  in  $\text{PM}_{2.5}$  as oil combustion or  
30 industry as their interpretation of the source (Viana et al., 2008), dependent on the area

1 surrounding the site. With an average contribution of 17% on an annual basis, this factor does  
2 not change significantly over the seasons. The SW, NE and INT.1 monsoons have roughly the  
3 same percentage of around 16-17%. INT.2 however scores the highest at 24% (V/Ni ratio =  
4 2.36), triple the HAZE events at only 7% (V/Ni ratio = 1.74). The slight inconsistencies of the  
5 percentage portion seasonally may be due to different batches of heavy oil and origins of  
6 crude oil, as discussed by Jiang et al. (2014) based on studies by Mueller et al. (2011) and  
7 Zaki et al. (1989).

8 Factor 1 seems to not be particularly affected by gaseous parameters or meteorological  
9 conditions, as reported in Table S4. Overall, API and this factor did not correlate well, with an  
10 exception during NE ( $r = 0.366$ ;  $p = 0.047$ ). WD is the only meteorological parameter that is  
11 significantly correlated with this factor, and this occurred during SW ( $r = 0.581$ ;  $p = 0.007$ )  
12 which may have resulted from HAZE ( $r = 0.677$ ;  $p = 0.045$ ). For gaseous parameters, factor 1  
13 seemed to correlate with gaseous parameters mostly during the NE monsoon, with significant  
14 positive correlations with CO ( $r = 0.498$ ;  $p = 0.005$ ), SO<sub>2</sub> ( $r = 0.436$ ;  $p = 0.016$ ), NO<sub>x</sub> ( $r =$   
15  $0.471$ ;  $p = 0.009$ ) and NO<sub>2</sub> ( $r = 0.529$ ;  $p = 0.003$ ). O<sub>3</sub> is the only gas that appears to have more  
16 than one season correlating significantly with this factor. A negative correlation was shown  
17 between this factor and O<sub>3</sub> during SW ( $r = -0.605$ ;  $p = 0.001$ ), while a positive correlation ( $r =$   
18  $0.796$ ;  $p = 0.032$ ) was seen during INT.2. Annually, only O<sub>3</sub> and SO<sub>2</sub> have significant  
19 correlations with this factor at  $r = -0.287$  ( $p = 0.014$ ) and  $r = 0.380$  ( $p = 0.001$ ), respectively.  
20 The positive correlation during INT.2 was perhaps due to higher measurements of NO<sub>x</sub> and  
21 NO<sub>2</sub> during this time period compared to other seasons. NO<sub>2</sub> provides an O-radical which  
22 contributes to the formation of O<sub>3</sub> with the assistance of sunlight and volatile organic  
23 compounds (VOCs). High concentrations of O<sub>3</sub> and other organic pollutants can lead to the  
24 formation of secondary organic aerosol; this may explain the observation results.

### 25 **3.3.2 Factor 2: mineral dust (Al, Li, U, Fe, Co, Ca<sup>2+</sup>, Sr, Mn, Mg<sup>2+</sup>)**

26 Factor 2 makes up 14% of the PM<sub>2.5</sub> mass (annual average). This factor was identified based  
27 on elements Al (77% of the Al mass), Li (61% of the Li mass), U (45% of the U mass), Fe  
28 (40% of the Fe mass), Co (38% of the Co mass), Ca<sup>2+</sup> (33% of Ca<sup>2+</sup> mass) and Mg<sup>2+</sup> (28% of  
29 Mg<sup>2+</sup> mass), as shown in Fig. 4a. Researchers cite these elements as markers for a mineral  
30 dust source. For example, Al and Fe were cited by Viana et al. (2008), Li and Fe by Pey et al.  
31 (2009) while Al and Fe by Balakrishnaiah et al. (2012). Mustaffa et al. (2014) reported a

1 mineral dust source based on the presence of  $\text{Ca}^{2+}$  while Zhang et al. (2011) have used  $\text{Mg}^{2+}$   
2 and  $\text{Ca}^{2+}$  as the indicators for a mineral dust factor.  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  were also used to classify  
3 crust ions in  $\text{PM}_{2.5}$  (Wang et al., 2005). Fe also represents typical soil constituents and/or  
4 crustal combustion (Ho et al., 2006; Aldabe et al., 2011).

5 During three consecutive seasons of the year, i.e. the SW, INT.2 and NE monsoons (middle  
6 May 2011 until early March 2012), the mineral dust source portion remains about the same at  
7 around 15-16% of the  $\text{PM}_{2.5}$  mass. However, during the following inter-monsoon (INT.1), this  
8 factor was reduced to half at 7%. The HAZE events on the other hand recorded the highest  
9 portion of this source with 19% of the  $\text{PM}_{2.5}$  mass. The reason is shown from the relationship  
10 between this factor and meteorological factors during this time period. This factor during  
11 HAZE seems to be affected by a few gaseous parameters i.e.  $\text{NO}_x$  and  $\text{NO}$  with  $r = 0.650$  ( $p =$   
12  $0.042$ ) and  $r = 0.698$  ( $p = 0.025$ ), respectively. Annually, only  $\text{SO}_2$  and  $\text{NO}_2$  have significant  
13 relationships with factor 2,  $r = 0.345$  ( $p = 0.005$ ) and  $r = 0.260$  ( $p = 0.035$ ). Except during both  
14 inter-monsoons, mineral dust had a significant relationship towards T (strong positive  
15 correlation) and RH (strong negative correlation) including HAZE which happens during the  
16 SW monsoon. This may be the reason why the SW monsoon and factor 2 records the strongest  
17 correlation compared to other seasons in Malaysia, with  $r = 0.673$  ( $p < 0.001$ ) towards T and  $r$   
18  $= -0.734$  ( $p < 0.001$ ) towards RH.

### 19 **3.3.3 Factor 3: mixed SIA and biomass burning ( $\text{NH}_4^+$ , Se, $\text{K}^+$ , $\text{SO}_4^{2-}$ , Rb)**

20 The combined sum of ammonium sulfate and ammonium nitrate represents the secondary  
21 inorganic contribution to the  $\text{PM}_{2.5}$  mass. This study is clearly dominated by ammonium  
22 sulfate. The potassium ion ( $\text{K}^+$ ) on the other hand is an indication of major soil elements,  
23 usually from biomass burning. Echalar et al. (1995) has indicated that potassium (K) may be  
24 considered a good tracer for the flaming phase of forest fires. Watson and Chow (2001)  
25 reported that 85% of the K is in the soluble form  $\text{K}^+$ , which is consistent with most vegetative  
26 burning profiles. Due to this established relationship,  $\text{K}^+$  in PM was seen in many studies as a  
27 marker of biomass origin, either in the European region (Reisen et al., 2013) or the SEA  
28 region (Tahir et al., 2013b; Wahid et al., 2013; Mustaffa et al., 2014; Ee-Ling et al., 2015).  
29 Reche et al. (2012) reported that  $\text{K}^+$  from biomass burning was mostly emitted in the fine  
30 fraction of PM rather than coarse particles. Characterised by high levels of  $\text{NH}_4^+$  (59% of  
31  $\text{NH}_4^+$  mass),  $\text{SO}_4^{2-}$  (46% of  $\text{SO}_4^{2-}$  mass) and  $\text{K}^+$  (49% of  $\text{K}^+$  mass), the third and biggest factor



1 for this site was identified as a mix of SIA and biomass burning and makes up 42% of the  
2 PM<sub>2.5</sub> mass on annual basis. Studies by Mooibroek et al. (2011), Zhang et al. (2013), Almeida  
3 et al. (2005), Yin et al. (2010) and Song et al. (2006) also identified a major contribution by  
4 the secondary aerosol fraction to PM<sub>2.5</sub>.

5 In this study, highest mass contribution of factor 3 was observed during the SW monsoon  
6 (51%) during which haze episodes normally occur. The rest of the year i.e. INT.2, NE and  
7 INT.1 represent 35% or less of the PM<sub>2.5</sub> mass i.e. 35%, 34% and 26% respectively. Except for  
8 INT.1, the other seasons show very significant correlations between this factor and secondary  
9 aerosol components, i.e. SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and K<sup>+</sup>. During HAZE, this factor contributed 63% of  
10 the PM<sub>2.5</sub> mass. The time series (Fig. 5c) shows that this factor's elevated contribution  
11 occurred during a period from July until the end of October which is when the haze episodes  
12 normally occur. The HYSPLIT back trajectories analysis traced back the mass from the HAZE  
13 samples to Sumatra, i.e. Palembang during the 2011 episode and Palembang/Pekan Baru for  
14 2012 episode; Fig. S4 a(ii), b(ii). This strongly suggested that during the period of the SW  
15 monsoon, the mass contribution of SIA and biomass factor could originate from long-range  
16 transport (regional influence) in addition to local agricultural and/or anthropogenic activities.

17 As shown by the factor-gaseous-meteorological correlation results, this factor on an annual  
18 scale seems to not correlate well with meteorological parameters, except for API and T.  
19 Season-wise, only API correlated well with this factor during SW and INT.2. However, on an  
20 annual scale, gaseous parameters showed varied relationships. CO, O<sub>3</sub>, and NO<sub>2</sub> showed  
21 significant positive correlations towards this factor 3 while NO revealed a significant negative  
22 correlation. The strongest correlations between this factor and gaseous-meteorological  
23 parameters were observed during the SW monsoon season. With highest mass contribution  
24 during SW, this factor showed significant ( $p < 0.05$ ) positive correlations with CO, O<sub>3</sub>, and  
25 NO<sub>2</sub> at  $r = 0.612$ ,  $r = 0.597$  and  $r = 0.422$ , respectively. The HAZE events on the other hand,  
26 although normally occurring during the SW monsoon, did not share these relationships. This  
27 factor during HAZE only correlated strongly with WS ( $r = -0.678$ ;  $p < 0.05$ ).

#### 28 **3.3.4 Factor 4: mixed traffic and industrial (NO<sub>3</sub><sup>-</sup>, Pb, NO<sub>2</sub><sup>-</sup>, Zn, As, Bi, Cd,** 29 **BC)**

30 Dominated by NO<sub>3</sub><sup>-</sup> (69% of NO<sub>3</sub><sup>-</sup> mass), Pb (58% of Pb mass), NO<sub>2</sub><sup>-</sup> (58% of NO<sub>2</sub><sup>-</sup> mass), Zn  
31 (55% of Zn mass), As (51% of As mass), Bi (47% of Bi mass), Cd (44% of Cd mass) and BC

1 (38% of BC mass), factor 4 was identified as mixed traffic and industrial sources with an  
2 average contribution of 10% on an annual scale. As shown in Table 4 and illustrated in Fig.  
3 5c, this factor varied with changing seasons. High contributions were seen from middle  
4 September until March during INT.2 (19%) and NE (20%) and very low contributions were  
5 seen during SW (4%) and INT.1 (6%) from April until September. HAZE appears to not to  
6 have significantly contributed to this factor with only 3% mass contribution. Most of the trace  
7 elements in this factor are related to both traffic (Pb, Zn) and industrial emissions (As, Ni)  
8 (Fang et al., 2003; Querol et al., 2007). Pb and Zn are enriched in both vehicular emissions  
9 and also industrial emissions (Song et al., 2006; Wåhlin et al., 2006; Querol et al., 2008; Pey  
10 et al., 2009; Thurston et al., 2011; Srimuruganandam and Shiva Nagendra, 2012b, a). EF  
11 results further suggest the Pb, Zn, As, Cd and Bi originated from anthropogenic sources.  
12 Malaysia has banned the use of Pb in petrol since 1996, indicating that the element is not  
13 originating from leaded petrol vehicle emissions. Thus, we exclude the influence of leaded  
14 petrol on this factor. Pastuszka et al. (2010) explain Pb mass as re-suspended road dust while  
15 Heal et al. (2005) explain Pb as road traffic emissions. Ewen et al. (2009) suggested that apart  
16 from the wear and tear of tyres, Cd is mainly emitted from the combustion of diesel fuel and  
17 oil or lubricants. Arsenic (As) mainly comes from industrial sources (Sánchez de la Campa et  
18 al., 2008; Stortini et al., 2009). Additionally, BC is an established tracer for primary  
19 anthropogenic emissions where its variability reflects changes in source strength, long-range  
20 transport and atmospheric mixing characteristics (Viidanoja et al., 2002). BC also is a major  
21 component of the PM<sub>2.5</sub> associated with road traffic emissions (Richmond-Bryant et al., 2009;  
22 Doumbia et al., 2012) and fuel oil combustion (Meyer, 2012; Zheng et al., 2012). Park et al.  
23 (2002) reported that the varying traffic and meteorological conditions of a site as well as the  
24 distance of the sampling equipment from the road traffic source will strongly influence the BC  
25 concentration. Data from the Malaysian Institute of Road Safety Research (MIROS) recorded  
26 a total of 342,279 vehicles in 24 h for the Federal Highway in October 2011 (Ministry of  
27 Works, 2011) which is near to our sampling station. During the peak hour of 0800 to 0900,  
28 24,016 vehicles were recorded on this road. Previous studies have shown that road traffic can  
29 make substantial contributions to particulate mass concentrations in the Klang Valley area  
30 (Awang et al., 2000; Afroz et al., 2003; Rahman et al., 2011; Abdullah et al., 2012; Wahid et  
31 al., 2013; Ee-Ling et al., 2015; Khan et al., 2015b). NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup> could also possibly come  
32 from the secondary aerosol of ammonium nitrate from anthropogenic activities in the

1 surrounding area such as motor vehicle exhaust, industries (petrochemical industry, iron/steel  
2 plant, etc), and stationery combustion sources (coal plants, etc).

3 With  $\text{NO}_3^-$  and  $\text{NO}_2^-$  ions in the factor, a relationship between this factor and gaseous elements  
4 is anticipated. On an annual scale, NO, NO<sub>x</sub> and NO<sub>2</sub> have shown significant positive  
5 correlations with this factor with  $r = 0.428$  ( $p < 0.001$ ),  $r = 0.459$  ( $p < 0.0001$ ), and  $r = 0.311$   
6 ( $p = 0.008$ ) respectively, indicating a traffic emissions source. WS showed a significant  
7 negative relationship with this factor ( $r = -0.39$ ;  $p < 0.001$ ). Season-wise, following high mass  
8 contribution, this factor during INT.2 showed significant correlation with NO<sub>x</sub> and NO<sub>2</sub> with  $r$   
9  $= 0.774$  ( $p < 0.05$ ) and  $r = 0.766$  ( $p < 0.05$ ), respectively. On the other hand, during the NE dry  
10 season, this factor showed a negative correlation with O<sub>3</sub> ( $r = -0.351$ ;  $p < 0.05$ ) and WS ( $r =$   
11  $-0.507$ ;  $p < 0.05$ ). Beckerman et al. (2008) reported that even though the level of NO<sub>2</sub> decay  
12 increases with increasing distance from the highway (at ~300 m), PM<sub>2.5</sub> still correlated  
13 strongly ( $r > 0.7$ ;  $p < 0.05$ ) with NO<sub>2</sub>, NO and NO<sub>x</sub>. They also found out that NO<sub>2</sub> still shows  
14 a strong association with PM<sub>2.5</sub> even with the potential of meteorological influences on the  
15 correlations. Pey et al. (2009) identified vehicle exhaust emissions based on high loadings of  
16 NO and CO in the principle components. A study in Korea by Park et al. (2002) concluded  
17 that BC is strongly correlated with CO and NO<sub>x</sub> which can be further used as a vehicle  
18 emission tracer for the Seoul urban area. In addition, they also found that a PM<sub>2.5</sub>-BC  
19 regression towards WS was negative, which is similar to our findings. These arguments  
20 further confirm the significance of our source type.

### 21 **3.3.5 Factor 5: sea salt (Na<sup>+</sup>, Cl<sup>-</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>)**

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

1 The understanding of the sea salt contribution during INT.1 requires some extended analysis.  
2 To investigate this, we carried out further stoichiometric analysis on the selected elements.  
3 The ratio of  $Mg^{2+}/Ca^{2+}$  on an annual scale was 0.11 while the seasonal ratios were: SW = 0.  
4 10, INT.2 = 0.083, NE = 0.072 and INT.1 = 0.24. The  $Cl^-/Na^+$  ratios for all seasons were: SW  
5 = 0.11, INT.2 = 0.056 and NE = 0.14 and INT.1 = 0.041, with an overall annual ratio of  
6 0.057. From these results, it is obvious that INT.1 contributed more  $Ca^{2+}$  and  $Na^+$  with higher  
7 occurrences of chloride loss or the “chlorine deficiency” phenomenon compared to other  
8 seasons. According to Song and Carmichael (1999), chlorine in fine particles is almost  
9 exhausted in just 24 h. Khan et al. (2010b) have reported that Cl loss in their study area is due  
10 to high humidity. Since Peninsular Malaysia is at the equator with very high T and RH,  
11 “chlorine deficiency” is a valid explanation. A similar observation of a low  $Cl^-/Na^+$  ratio was  
12 also reported for Kuala Terengganu, Malaysia, at 0.02 (Tahir et al., 2013b). The  $PM_{2.5}$  marine  
13 influence towards the sea salt factor has also been discussed elsewhere (Almeida et al., 2005,  
14 2006).

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

### 21 **3.3.6 HAZE**

22 As shown in Fig. 2a, two haze episodes occurred during our sampling period. The first  
23 episode occurred in September 2011 during the SW monsoon and the second episode  
24 occurred in June 2012, also during the SW monsoon. Since both episodes occurred during the  
25 same season, it is anticipated that both episodes have similar characteristics and therefore  
26 share the same origin. However, our investigation, as reported in Table S5, revealed that the  
27 two episodes to have quite different characteristics. Although both episodes were most  
28 strongly influenced by the same source of mixed SIA and biomass burning, other sources did  
29 not follow the same trend. For a total of 19% during HAZE 2011, four other factors were  
30 identified: combustion of engine oil, sea salt, mineral dust, and mixed traffic and industrial.  
31 These factors do not seem to have a strong influence on HAZE 2011. However, HAZE 2012

1 was strongly influenced by those four factors, with a combined contribution of 44%. As  
2 reported in Table S6, the PMF factor 3 of mixed SIA and biomass burning was further  
3 investigated through a correlation matrix between CMC and the source for a better  
4 understanding of the composition/characteristics. HAZE 2012 showed a significant  
5 correlation between PMF factor 3 (mixed SIA and biomass burning) and CMC SIA with  $r =$   
6  $0.952$ ,  $p < 0.001$ . The PMF factor 3 during HAZE 2012 also showed significant correlations  
7 ( $p < 0.001$ ) with  $\text{SO}_4^{2-}$  ( $r = 0.963$ ),  $\text{NH}_4^+$  ( $r = 0.944$ ) and  $\text{nss-SO}_4^{2-}$  ( $0.965$ ) but not with  $\text{K}^+$ .  
8 Further, the CMC SIA showed significant correlations with  $\text{SO}_4^{2-}$  ( $r = 0.995$ ;  $p < 0.0001$ ),  
9  $\text{NH}_4^+$  ( $r = 0.997$ ;  $p < 0.0001$ ) and  $\text{K}^+$  ( $r = 0.829$ ;  $p = 0.011$ ). Therefore, we could conclude that  
10 PMF factor 3 (mixed SIA and biomass burning) during HAZE 2012 was in fact influenced by  
11 both SIA and biomass burning. HAZE 2011, however, indicated different sources. The PMF  
12 factor 3 did not have any significant correlation with CMC SIA, any of the CMC SIA  
13 elements or  $\text{K}^+$ . However, CMC SIA showed significant correlation with CMC  $\text{SO}_4^{2-}$  ( $r = 1$ ;  $p$   
14  $= 0.016$ ) and CMC  $\text{NH}_4^+$  ( $r = 1$ ;  $p = 0.02$ ) but no significant correlation towards  $\text{K}^+$ . These  
15 results indicate that HAZE 2011 was mostly influenced by SIA alone and less so by biomass  
16 burning. With 10% mass contribution from combustion of engine oil, HAZE 2011 could be  
17 concluded to have been influenced by anthropogenic activities including traffic. Besides SIA,  
18 a significant influence of mineral dust (25%) and sea salt (9%) showed that HAZE 2012 was  
19 greatly influenced by long-range transport. HYSPLIT backward trajectories for both HAZE  
20 episodes were traced back to Sumatra, Indonesia; Fig. S4 a(ii), b(ii). Further analysis showed  
21 that HAZE 2012 was more influenced by the meteorological and gaseous parameters whereas  
22 during HAZE 2011, strong correlations were observed but they are not significant; as shown  
23 in Table S7. However, it is still not clear whether long-range transport did impact our HAZE  
24 episodes.

### 25 **3.4 Comparison between CMC and PMF Source**

26 As shown in Fig. 4b and Fig. S5, predicted mass modelled by PMF and reconstructed mass by  
27 CMC were compared to those measured  $\text{PM}_{2.5}$  mass. Both approaches resulted with good  
28 regression at  $R^2 = 0.901$  and  $R^2 = 0.784$ , respectively. Further, seasonal regressions and time  
29 series between these two approaches were shown in Fig. S6. The analysis were run on  
30 selected components that have similarity to compare: 1) CMC dust vs. PMF factor 2 mineral

1 dust; 2) CMC SIA and  $K^+$  vs. PMF factor 3 SIA and biomass burning; and 3) CMC sea salt vs.  
2 PMF factor 5 sea salt.

3 Overall, 'dust' and 'SIA and biomass' component shows similar trend, as shown in Fig. S6.  
4 Both component have good correlations between CMC and PMF approach ( $R^2$  more than  
5 0.70) except during INT.1 and NE monsoon ( $R^2$  less than 0.50). Rainfall, which was higher  
6 during these two seasons compared to other season, could be the reason. The ANNUAL  
7 CMC/PMF ratio for seasonal 'dust' component is 0.29 ( $r = 0.89$ ) while seasonal regression  
8 (including HAZE) range between 0.24 and 0.53. The seasonal CMC/PMF ratio for 'SIA &  
9 biomass' component ranges between 0.13 and 0.24 with an annual ratio of 0.15 ( $r = 0.87$ ). The  
10 inconsistency ratio for both components maybe due to the fact that PMF contribution includes  
11 absorbed elements other than selected elements considered by the CMC approach. The 'sea  
12 salt' component however, did not show good agreement between two approaches. However,  
13 for HAZE dataset, the two approaches seem to have a very good agreement ( $r = 0.94$ ) on 'sea  
14 salt' component. As shown in Fig. S6c, the seasonal regression of CMC and PMF for sea salt  
15 showed that this pair is moderately correlated. Similar observation of large difference on 'sea  
16 salt' component (also known as marine aerosol) between two approaches with one of them  
17 being the mass closure calculation was seen and discussed by Almeida et al. (2006) and  
18 Farao et al. (2014). The observation on this matter was that perhaps CMC calculation did not  
19 include all the components associated with sea spray and due to reaction of NaCl with  
20 inorganic acids ( $HNO_3$  and  $H_2SO_4$ ) which resulted with the loss of  $Cl^-$  ion.

21 The different estimation derived from the two approaches was expected. According to  
22 Harrison et al. (2003), CMC is a hybrid between comprehensive chemical analysis method  
23 and simpler statistical procedure. It is a simple approach yet effective model to assort the  
24 measured PM compounds into different source categories. One of the highlights of this  
25 method is that CMC treats sulphate and nitrate separately. This is crucial since different  
26 ambient condition can leads to different response of the aerosol which will further affect  
27 organic carbon thus secondary organic carbon trend. Moreover, CMC also separate the sea salt  
28 and crustal components which have different respond to changes in traffic volume. PMF on  
29 the other hand, is an advanced computational tool to identify sources and eventually the mass  
30 contribution based on the work by Paatero and Tapper (1994). So, it is likely to have two  
31 different results and thus almost impossible to verify results from the two different methods  
32 (Hellén et al., 2003; Hopke et al., 2006; Vallius et al., 2008; Vecchi et al., 2008; Favez et al.,

1 2010; Hellebust et al., 2010). This issues have been highlights by Viana et al. (2008) on  
2 Europe source apportionment studies. The study stated that it is difficult to obtain coinciding  
3 results with different receptor models for the same data. This statement is supported by Vallius  
4 et al. (2008) which stated that different methods yield different results when they are applied  
5 to air pollution data.

6

#### 7 **4 Conclusions**

8 Our results revealed that fine particles are very significant in the ambient air of the Petaling  
9 Jaya urban-industrial area in the Klang Valley. The PM<sub>2.5</sub> mass averaged  $28 \pm 18 \mu\text{g m}^{-3}$   
10 which is almost triple (2.8 fold) the WHO annual guideline. Our result is higher than reported  
11 for other parts of Peninsular Malaysia, but very low compared to other large Asian cities and  
12 variable when compared to other parts of the world. On a daily basis, the PM<sub>2.5</sub> mass ranged  
13 between 6 to  $118 \mu\text{g m}^{-3}$  with 43% (samples) exceedance of the daily WHO guideline. On  
14 average, weekends recorded lower PM<sub>2.5</sub> mass ( $26 \mu\text{g m}^{-3}$ ) compared to weekdays ( $29 \mu\text{g m}^{-3}$ ).  
15 The month of June during the dry season of the SW monsoon recorded the highest monthly  
16 average at  $61 \mu\text{g m}^{-3}$  while November during the wet season of the NE monsoon recorded the  
17 lowest ( $17 \mu\text{g m}^{-3}$ ). The NE monsoon is the only season that did not have more than 50%  
18 exceedance of the daily WHO guideline.

19 In relation to meteorological-gaseous parameters, PM<sub>2.5</sub> mass on an annual scale showed the  
20 strongest relationship with API ( $r = 0.763$ ;  $p < 0.001$ ), explained by the PM<sub>2.5</sub>/PM<sub>10</sub> ratio  
21 (0.72). As anticipated, PM<sub>2.5</sub> was positively correlated with T and WS but negatively  
22 correlated with RH. Rainfall and WD were not found to be significantly influential. With an  
23 exception of NO<sub>x</sub>, all other gaseous parameters were found to significantly influence the  
24 PM<sub>2.5</sub> mass. CO, NO<sub>2</sub> and SO<sub>2</sub> were found to significantly correlate with PM<sub>2.5</sub> indicating a  
25 combustion-related traffic source. NO was the only gaseous parameter that had a negative  
26 relationship with PM<sub>2.5</sub> mass. O<sub>3</sub> at the site was also significantly correlated with PM<sub>2.5</sub> mass.

27 On a seasonal scale, daily PM<sub>2.5</sub> mass in all seasons was affected by the gaseous parameters  
28 but not meteorological conditions. The SW monsoon was found to have a significant  
29 relationship with CO, O<sub>3</sub>, NO<sub>2</sub> and API while the NE monsoon was correlated with SO<sub>2</sub> and  
30 NO<sub>2</sub>. Having relatively uniform T, RH and copious rainfall throughout the year, the small  
31 influence of meteorological parameters towards seasonal PM<sub>2.5</sub> mass variation was as

1 anticipated. All four seasons showed positive correlations with PM<sub>2.5</sub> mass and T but the  
2 HAZE events revealed a slight negative correlation. The RH and PM<sub>2.5</sub> relationship was  
3 negative except during INT.1. Unexpectedly, rainfall, WS and WD did not significantly  
4 correlate with PM<sub>2.5</sub> mass variability with changing season even during the major seasons of  
5 the SW or NE monsoons. Further analysis on the PM<sub>2.5</sub>/PM<sub>10</sub>, PM<sub>2.5</sub>/TSP and PM<sub>10</sub>/TSP ratios  
6 revealed that meteorological parameters only greatly influence the coarse particles (particle  
7 with an aerodynamic diameter of greater than 2.5 μm) but not so much on fine particles  
8 (particle with an aerodynamic diameter of less than 2.5 μm) at this site.

9 The PM<sub>2.5</sub> chemical compositions determined were anions, cations, TE and BC for a total of  
10 36% of the PM<sub>2.5</sub> mass. The total cation to total anion ratio was 0.46 with the ions in the  
11 decreasing trend: SO<sub>4</sub><sup>2-</sup> > NH<sub>4</sub><sup>+</sup> > Na<sup>+</sup> > K<sup>+</sup> > NO<sub>3</sub><sup>-</sup> > Ca<sup>2+</sup> > PO<sub>4</sub><sup>3-</sup> > Mg<sup>2+</sup> > Cl<sup>-</sup> > Br<sup>-</sup> > NO<sub>2</sub><sup>-</sup>  
12 > F. TE analysis revealed Al, Fe, Zn, and Pb as the major elements. It is notable that results  
13 for Pb, As, Cd and Ni in this study did not exceed any EU standard on air pollutants. We  
14 further constructed CMC to better understand the seasonality variability in PM<sub>2.5</sub> composition.  
15 Our finding showed that both primary and secondary components of PM<sub>2.5</sub> are equally  
16 important, albeit with seasonal variability. The CMC components identified were: BC > SIA >  
17 Dust > TE > Sea salt > K<sup>+</sup>. Seasonally, BC showed highest accountability during the NE  
18 monsoon and lowest during the SW monsoon but other CMC components did not vary largely  
19 with changing season. As for the SIA, the NR of 0.26 indicated an excess of SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> at  
20 the site. Further SIA components analysis revealed that SIA at the site was affected by  
21 anthropogenic activities rather than marine influences. The EF analysis further distinguished  
22 trace elements into two groups from anthropogenic sources (Pb, Se, Zn, Cd, As, Bi, Ba, Cu,  
23 Rb, V and Ni) and crustal sources (Sr, Mn, Co, and Li).

24 For SA purposes, we incorporated PMF 5.0 and MLR which revealed strong and significant  
25 correlations between the predicted and measured mass of PM<sub>2.5</sub> (R<sup>2</sup> = 0.901). Five factors  
26 were identified: 1) Combustion of engine oil; 2) Mineral dust; 3) Mixed SIA and biomass  
27 burning; 4) Mixed traffic and industrial; and 5) Sea salt; with an annual mean contribution of  
28 17, 14, 42, 10 and 17%, respectively. The dominance of each identified source largely varied  
29 with changing season but were roughly consistent with the CMC, EF and stoichiometric  
30 analysis for a few factors, accordingly. In addition to local anthropogenic activities, regional  
31 long-range transport was also influential. Further analysis on the HAZE episodes revealed  
32 different influences for the two different haze episodes. HAZE 2011 was mostly influenced by



1 SIA but not so much by biomass burning, indicating more influence from anthropogenic  
2 activities (including traffic). Meanwhile, HAZE 2012 could be greatly influenced by long-  
3 range transport with large contributions from SIA, biomass burning, mineral dust and sea salt.  
4 HYSPLIT backward trajectories for both HAZE episodes traced the air masses back to  
5 Sumatra, Indonesia.

6 These results are connected to the urban-industrial background of the area, where gaseous  
7 parameters affect  $PM_{2.5}$  mass both annually and seasonally. However, correlation between the  
8 chemical constituents and sources of  $PM_{2.5}$  towards meteorological and/or gaseous parameters  
9 largely varied with different season. Overall, this study suggests that  $PM_{2.5}$  and its constituents  
10 here in Klang Valley urban–industrial environment, were characterised by the local and  
11 regional activities as well as the seasonal tropical change. However, our study is limited to  
12 only fine particle. Parallel sampling of both fine and coarse particle will give better insight on  
13 the actual condition of the aerosol at a site. With the use of meteorological-gaseous  
14 parameters, concrete conclusion can be achieved, as to whether meteorological-gaseous  
15 parameters are affecting in the same way fine and coarse particles, and whether both fine and  
16 coarse particle share common sources. In addition to inorganic composition, further  
17 comprehensive assessment covering the organic portion and total elemental inorganic  
18 composition (i.e. total K, total Mg, total Na, total Ca, Si, S etc) is necessary for a complete  
19 composition dataset. In addition, it is suggested that particle number concentration (PNC)  
20 distribution should be incorporated into the chemical composition SA analysis as well. The  
21 potential source contribution function (PSCF) could also enhance the analysis of local and  
22 regional long-range transport. Alternatively, a simple yet effective approach, looking at the  
23 prevailing wind direction (or polar plots) associated to the different PMF factors could answer  
24 the effect of wind direction on PM. This would lead to improved analysis results and  
25 interpretation of the  $PM_{2.5}$  dataset, which eventually will lead to better understanding of the  
26 fine particle variability here in Klang Valley.

27

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10

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24

1 Table 1. Descriptive statistics of PM<sub>2.5</sub> mass and particulate matter (PM) ratio; unit: mean ± standard deviation (min - max). Remarks: SW =  
 2 South-west monsoon; NE = North-east monsoon; INT.2 = Inter-monsoon 2; INT.1 = Inter-monsoon 1; HAZE = samples with PM<sub>2.5</sub> mass more  
 3 than 40 µg m<sup>-3</sup> and air pollution index (API) more than 50.

	ANNUAL	SW	INT.2	NE	INT.1	HAZE
	5 Aug 2011 - 18 July 2012	15 May - 14 Sept	15 Sept - 30 Oct	1 Nov - 14 Mar	15 Mar - 14 May	
Elements	n = 81	n = 29	n = 7	n = 35	n = 10	n = 11
PM <sub>2.5</sub> (µg m <sup>-3</sup> )	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 <sub>2.5</sub> /PM <sub>10</sub>	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 <sub>2.5</sub> /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 <sub>10</sub> /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

4

1 Table 2. Comparison of PM<sub>2.5</sub> mass recorded in this study with other previous studies.

Location	PM <sub>2.5</sub> mass (ug m <sup>-3</sup> )	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	Urban	Jan - Mar 2013	Ee-Ling et al. (2015)
	18 ± 3	Metropolitan		
	10 ± 4	Semi-urban		
		Rural		
Kuala Lumpur, Klang Valley, Malaysia	27 + 10	Urban	Jan 2004 - Dec 2008	Rahman et al. (2011)
Kuala Terengganu, Malaysia	14 ± 7	Coastal, Sub-urban	Aug 2006 - Dec 2007	Tahir et al. (2013b)
Petaling Jaya, Klang Valley, Malaysia	33	Urban - industrial	Dec 1998 - Dec 2000	Keywood et al. (2003)
Gombak, Klang Valley, Malaysia	28	Urban - residential	Dec 1998 - Dec 2000	
New Taipei City, Taiwan	22 ± 8	Urban - Industrial	May 2011 - Nov 2011	Gugamsetty et al. (2012)
Agra, India	140 ± 22	Urban - Industrial	Nov 2010 - Feb 2011	Pachauri et al. (2013)
	308 ± 52	Traffic		
	91 ± 17	Rural		

Paris, France	15 + 10	Urban	11 Sept 2009 - 10 Sept 2010	Bressi et al. (2013)
	15 + 11	Semi-urban		
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	Urban	Mar 2009 - Jan 2010	Squizzato et al. (2013)
	33	Industrial		
	26	Semi-urban		
Birmingham, United Kingdom	12	Urban	May 2007 - Apr 2008	Yin et al. (2010)
	10	Rural		
Palermo, Sicily, Italy	34	Metropolitan; Urban 1	Nov 2006 - Feb 2008	Dongarrà et al. (2010)
	24	Urban 2		
Singapore	27 ± 10	Urban	Jan – Dec 2000	Karthikeyan and Balasubramanian (2006)

1 Table 3. Pearson correlation matrix results between seasonal PM<sub>2.5</sub> mass and: a)  
 2 meteorological; and b) gaseous parameters. Remarks: For meteorological parameters, API is  
 3 Air Pollution Index; T = temperature; RH = relative humidity; WS = wind speed; and WD =  
 4 wind direction.

a) Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
API	<b>0.763<sup>b</sup></b>	<b>0.748<sup>b</sup></b>	0.299	<b>0.473<sup>a</sup></b>	<b>0.705</b>	0.531
T	<b>0.310</b>	0.236	0.572	0.201	0.030	-0.050
RH	<b>-0.314<sup>a</sup></b>	-0.252	-0.495	-0.174	0.152	0.108
WS	<b>0.274</b>	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

5

6

b) Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
CO	<b>0.471<sup>b</sup></b>	<b>0.687<sup>b</sup></b>	0.713	<b>0.488<sup>a</sup></b>	<b>0.654</b>	<b>0.749<sup>a</sup></b>
O <sub>3</sub>	<b>0.298<sup>a</sup></b>	<b>0.535<sup>a</sup></b>	0.427	<b>0.433</b>	0.378	0.449
SO <sub>2</sub>	<b>0.324</b>	0.141	-0.250	<b>0.654<sup>b</sup></b>	0.627	0.445
NO <sub>x</sub>	0.058	0.112	<b>0.800</b>	<b>0.380</b>	0.588	0.192
NO	<b>-0.262</b>	-0.309	0.701	0.086	-0.126	-0.285
NO <sub>2</sub>	<b>0.473<sup>b</sup></b>	<b>0.528<sup>a</sup></b>	<b>0.851</b>	<b>0.711<sup>b</sup></b>	<b>0.874<sup>a</sup></b>	0.599

7

8 Values in bold are different from zero with a significance level alpha =0.05;

9 <sup>a</sup> is when p-values < 0.001 and

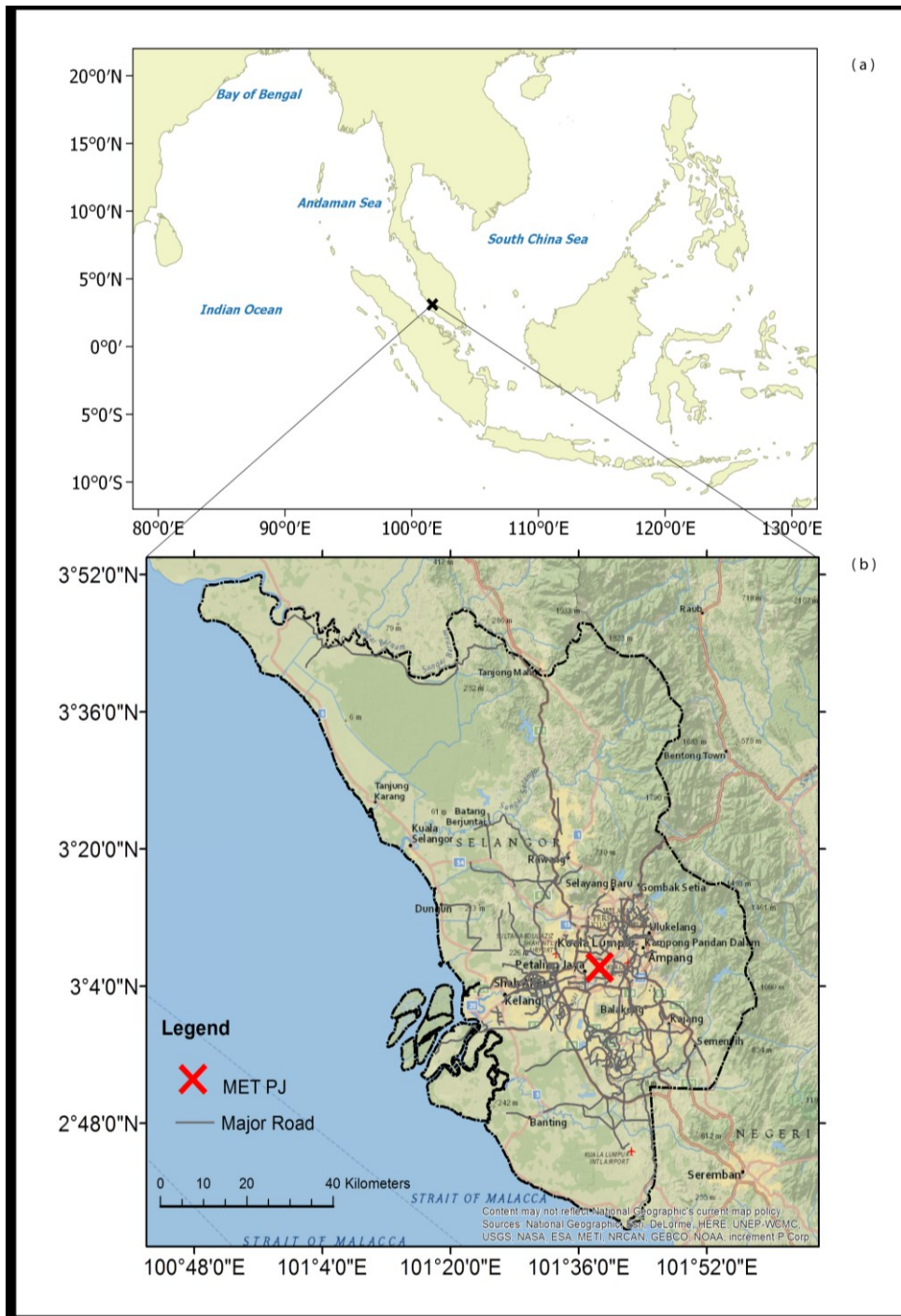
10 <sup>b</sup> p-values < 0.0001

11

1 Table 4. Relative contribution of PM<sub>2.5</sub> sources from the positive matrix factorisation (PMF) analysis. Remarks: SIA = secondary inorganic  
 2 aerosol.

Source contribution, $\mu\text{g m}^{-3}$ (%)	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%)

3



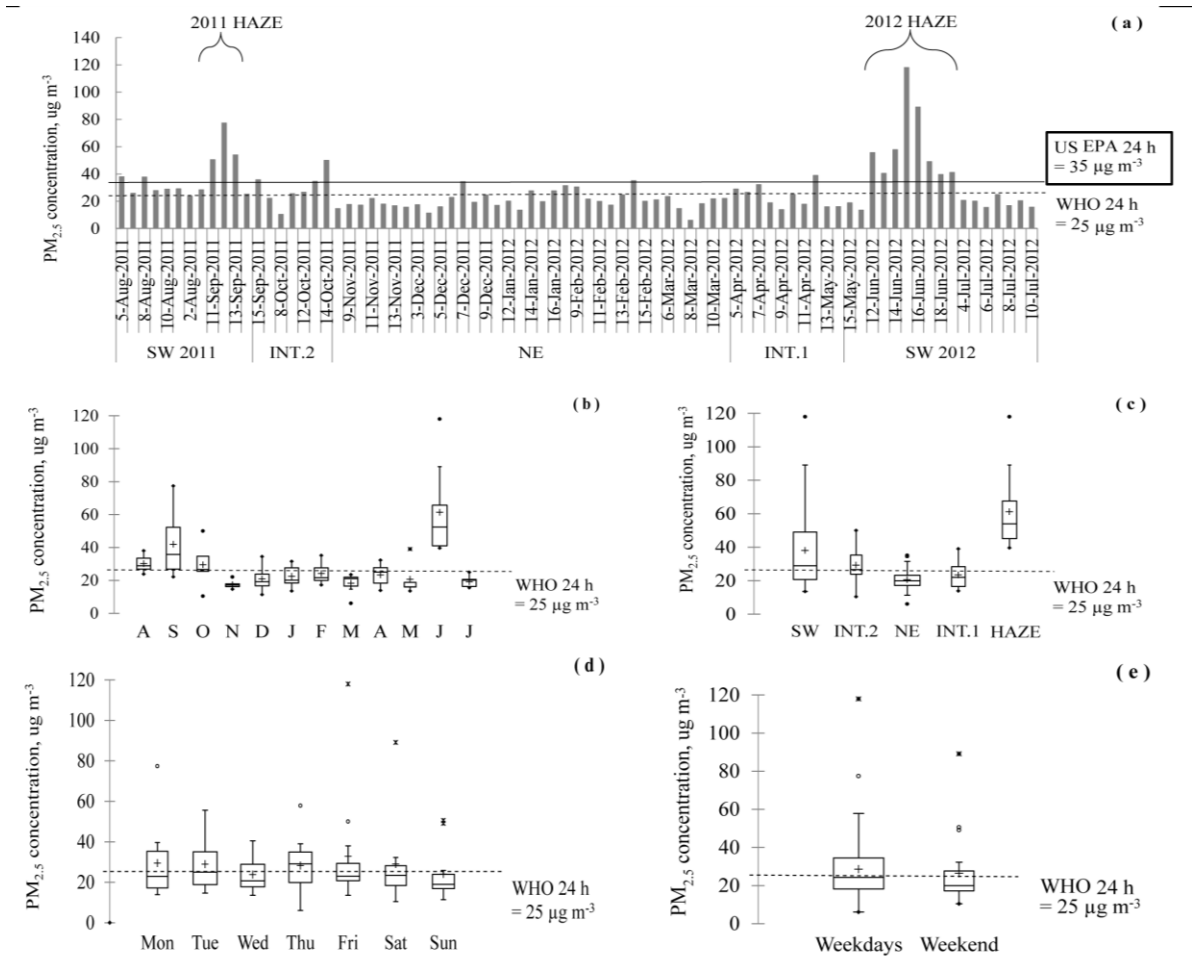
1

2

3 Figure 1. Location of the sampling site mark as “X” in: a) the Southeast Asia region; and b)

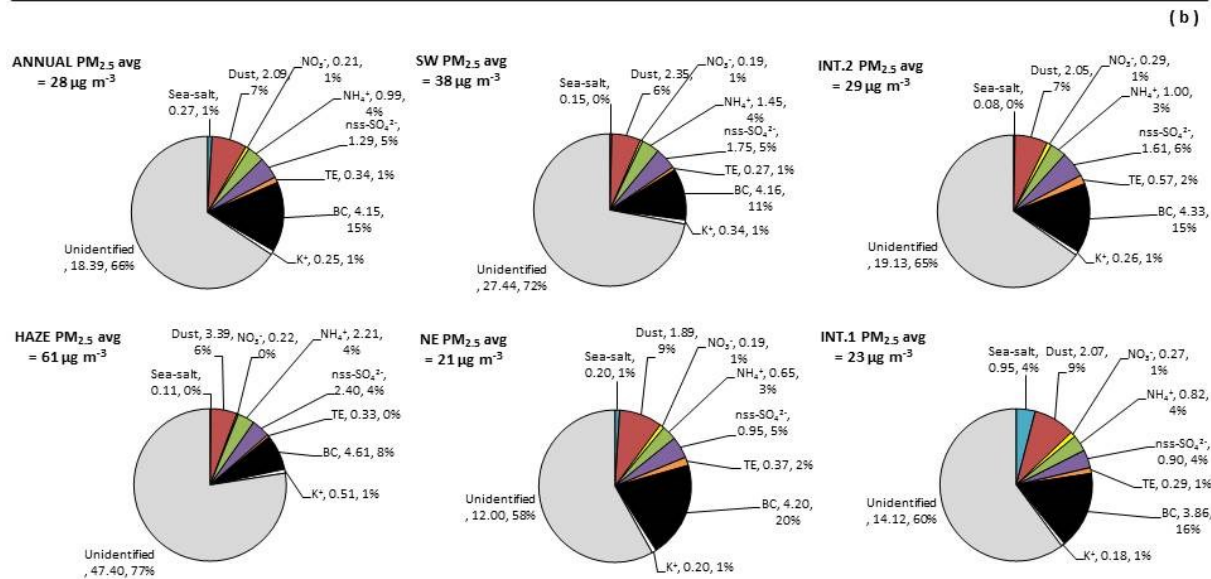
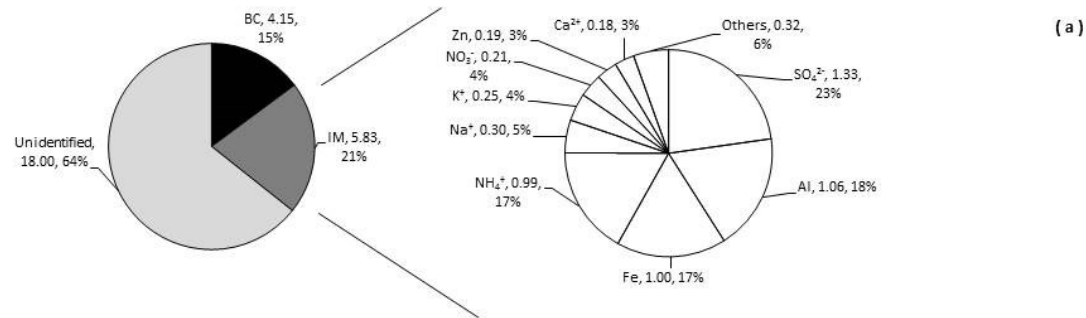
4 the Klang Valley area in the Peninsular Malaysia.





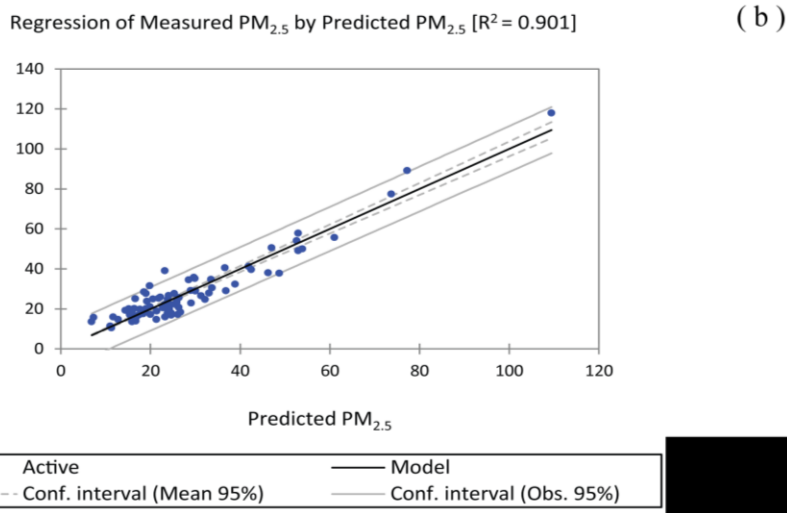
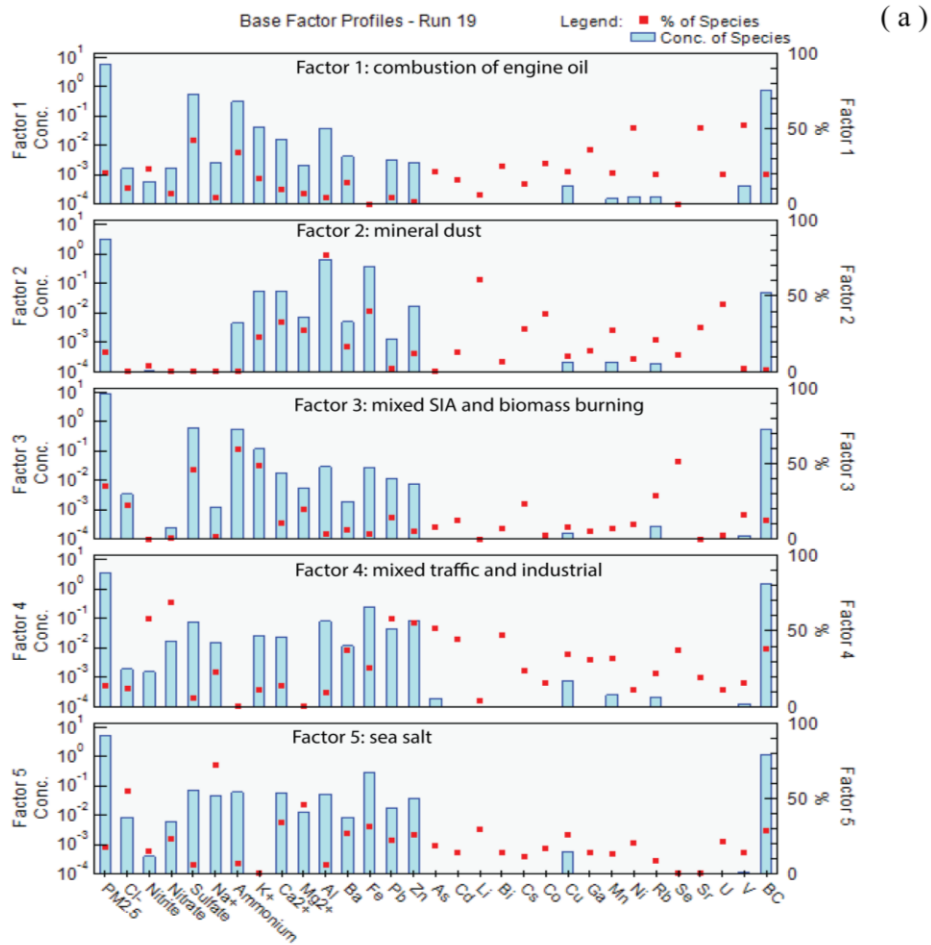
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3 Figure 2. The PM<sub>2.5</sub> mass concentration on the: (a) daily basis; with box and whisker plots (of  
 4 the: (b) monthly; (c) seasonal; (d) days; and (e) weekdays/weekend. For the box and whisker  
 5 plots, the horizontal line within the box indicates the median, boundaries of the box indicate  
 6 the 25<sup>th</sup> and 75<sup>th</sup> percentile, and the whiskers indicate the highest and lowest values of the  
 7 results. The “+” marked in the box indicates the mean. All figures were also subject to World  
 8 Health Organisation (WHO) daily PM<sub>2.5</sub> guideline and United States Environmental  
 9 Protection Agency (US EPA) daily PM<sub>2.5</sub> standard, accordingly.



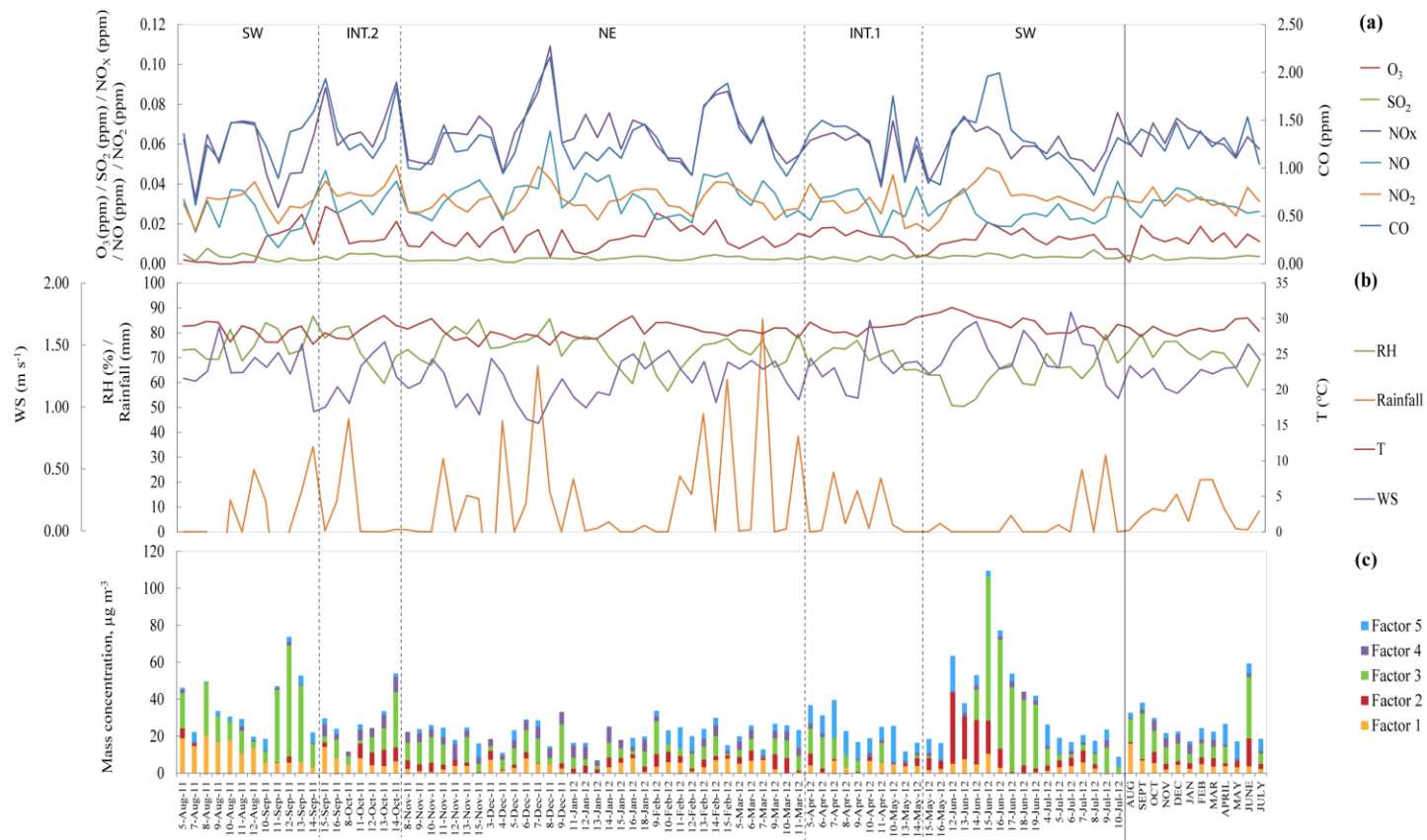
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3 Figure 3. The composition of PM<sub>2.5</sub> displayed as [element; mass in µg m<sup>-3</sup>; percentage in PM<sub>2.5</sub> mass] based on: a) annual chemical  
4 composition determined where IM is the inorganic matter; and b) seasonal chemical mass closure (CMC) components identified.



1

2 Figure 4. Source apportionment results from positive matrix factorisation (PMF) analysis: a)  
 3 source profile; and b) regression plot between measured and predicted  $PM_{2.5}$  mass. Remark:  
 4 SIA = secondary inorganic aerosol



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3 Figure 5. Time series of daily and monthly variations (left to right) of: a) gaseous; b) meteorological parameters; and c) mass concentration of  
 4 PM<sub>2.5</sub> sources.