

# Multivariate Analysis on Trace Elements of Fine Particulate Matter (PM<sub>2.5</sub>) in Industrial and Sub-Urban Area

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**Abstract:** Study on fine particles (PM<sub>2.5</sub>) and its effects towards health and environment has become as main concerned of present study. Therefore, the study was conducted to identify PM<sub>2.5</sub>, tracking the elements and sources at industrial and sub-urban area. E-BAM was used to measure the concentration of PM<sub>2.5</sub> and meteorological parameters during sampling. The PM<sub>2.5</sub> samples were analyzed to identify elements using inductively coupled plasma - optical emission spectrometry (ICP-OES). PM<sub>2.5</sub> concentrations were higher in industrial site (46.00 µg m<sup>-3</sup>) compared to sub-urban site (16.67 µg m<sup>-3</sup>), due to anthropogenic sources which were factories and major roads that located nearby study area. From PM<sub>2.5</sub> samples analysis, it showed that Al, K, and Ca elements were the highest average among 13 elements measured in PM<sub>2.5</sub> at both sites. Finally, based on PCA analysis, it showed that three main component groups had been identified: Na, Al, K, Ca, Ti, Sr, Cr and Mg can be associated with natural sources, Mn, Fe, Ni and Zn sources were linked to industrial activity sources, while Pb had been identified as a vehicle source for each area. In conclusion, the concentration of PM<sub>2.5</sub> and its elements in different locations provide different readings which depending on the sources of PM<sub>2.5</sub> at site. The finding from this study can assist local authorities to monitor and predict the highest concentration of PM<sub>2.5</sub> in problematic areas.

**Keywords:** anthropogenic activities; hysplit model; industrial emission; major pollutants; sub-urban emission;

## 1. Introduction

Physical and chemical characteristics of particulate matter (PM) including particle concentration, size distribution, and trace elements, which are defined from PM source, type and mixing processes in the atmosphere have been identified can give effect on climate and human health [1]. Recently, problems caused by atmospheric PM have not been limited to urban and industrial areas, but also can spread out to the entire regions. Every part of the world is connected through fast atmospheric transport, which allowed pollutants to travel at longer distance [2]. PM is small enough to remain airborne for an extended time, where it can be explained by hours or days which it can travel in considerable distances from the sources [3]. Researchers merely used two main PM size fractions that are PM<sub>10</sub> and PM<sub>2.5</sub>, where the aerodynamic diameters are less than 10 and 2.5 µm, respectively [4].

World Health Organization (WHO) highlighted the control of PM<sub>2.5</sub> for a more targeted response to the problems related to PM exposure [5]. WHO and USEPA has set the standard limit for 24 hr average of PM<sub>2.5</sub> concentration which are 25 µg m<sup>-3</sup> and 35 µg m<sup>-3</sup>, respectively, since 2005. Hence, in 2015, new Malaysia Ambient Air Quality Standard (MAAQS) has been introduced by Malaysian Department of Environment (DOE) with additional a new crucial air pollutants which is PM<sub>2.5</sub> for interim targets set including interim target (IT-1) in 2015, interim target 2 (IT-2) in 2018 and the full implementation of the PM<sub>2.5</sub> standard in 2020. For recent reference, interim target 2 (IT-2) become as guideline for both 24 h (50 µg m<sup>-3</sup>) and annually limit (35 µg m<sup>-3</sup>).

Sources of particles can be known by trace elements of PM<sub>2.5</sub>. Information of trace elements can be used both for qualitative identification of sources and quantification of contribution of such sources to the mass of PM<sub>2.5</sub>. Understanding on primary and secondary PM chemical data are very crucial. Most of the PM<sub>2.5</sub> studies in Malaysia are

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rather focus on mass concentrations of  $PM_{2.5}$  and no comparisons between sites such urban area, industrial area, sub-urban area, and reference area [6-11]. Physical and chemical characteristics of  $PM_{2.5}$  and temporal variation are important factors in determining the possible sources. In determination the possible sources, source apportionment are one of the approaches that can be applied. This approach can be divided into two procedures: i) identification of sources; and ii) apportionment of  $PM_{2.5}$  to the possible sources nearby the sampling location [12]. Principles, history and recent growths in atmospheric sciences on application of receptor modelling of PM has been reviewed by Viana et al. [12] and under the exploratory factor analysis models, commonly used model is principal component analysis (PCA) which based on the idea that time dependence of a chemical species at the receptor site will be similar to that other species from the similar sources or transport pathway [13]. According to Viana et al. [12], in Europe there are more than 30% of studies used PCA method in their source apportionment analysis up to year 2005. The advantage of using this method is simple yet efficient especially for exploratory source identification study. In addition, there is no specific software required and fast identification of source. Therefore, PCA should mainly be used for exploratory assessment as a preliminary result of the possible sources as used by many researchers [9]-[11], 14, 15].

Thus, the objective of this study was to identify possible sources of  $PM_{2.5}$  based on trace elements found at different site categories (i.e. industrial and sub-urban site) that affect the air quality such as industries, site construction dust, open burning and traffic emissions in the study area.

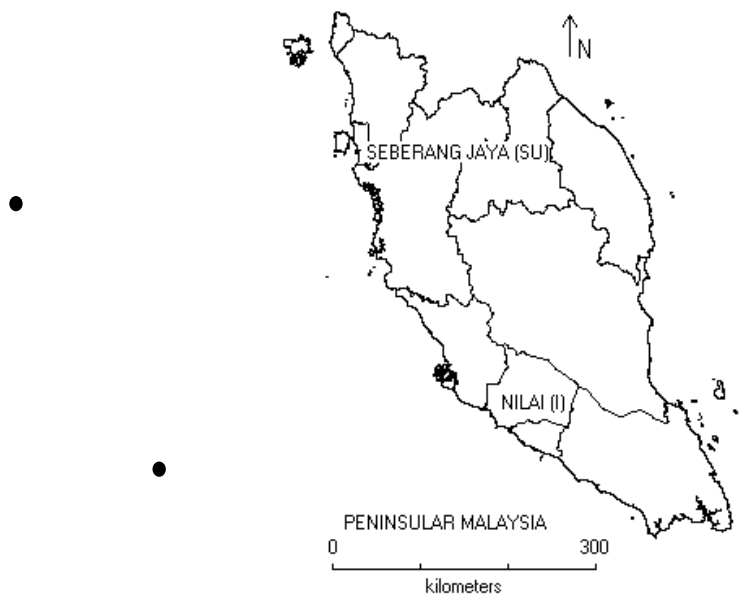
## 2. Methodology

### 2.1 Sampling Site Description

In this study, research was conducted at two (2) monitoring sites as represented industrial and sub-urban areas in Malaysia. The sampling sites within selected areas were assigned based on two criteria: (i) based on existing of continuous air quality monitoring station (CAQMS) done by Department of Environment, Malaysia (DOE); and (ii) based on location to reflect the  $PM_{2.5}$  emission sources through site profiling.  $PM_{2.5}$  data collection including the concentration and filter samples was set for 1 h average and conducted for 12 h per day for each site starting from 7 a.m. to 6 p.m. which during daytime only. This is due to safety factor for the instrument to be left on site for 24 h of operation. Descriptions of each sampling site was shown in Table 1 and locations of the selected sites were illustrated in Fig. 1.

**Table 1 - Specific details of monitoring location**

Category	Location	Date	Coordinate		Day	Sampling hour
Industrial (I)	Nilai, Negeri Sembilan	28 <sup>th</sup> Dec 2014- 3 <sup>rd</sup> Jan 2015	2°49'47.89"N	101°49'37.05"E	7	12
Sub-urban (SU)	Seberang Jaya, Pulau Pinang	16 <sup>th</sup> May 2016- 22 <sup>th</sup> May 2016	2°58'49.24"N	101°20'24.19"E	7	12



**Fig.1 - Location of selected sampling sites for Sub-urban (Seberang Jaya) and Industrial (Nilai) site**

Area profiling was done around site within 5 km radius from the monitoring station. Every possible source that had been found around the sampling site was recorded and the distance from monitoring stations was determined using a map obtained from Google map. In addition, all possible sources were marked on the map with a coordinate that obtained by using Global Positioning System (GPS) and record type of industry which those process was performed by travelling using a car. The information obtained from site profiling was combined with trace element analysis to investigate the possible sources that contributed to PM<sub>2.5</sub> based on wind direction at each site. PM<sub>2.5</sub> filter samples were collected hourly for each site and the hourly samples were used for digestion.

The industrial site is a rapidly developing town in Nilai, surrounded by many industrial areas and become the attraction center for investors in the industrial sector because of its strategic location, being adjacent to the Klang Valley (40km) and the Putrajaya Administrative Centre (38km) as well as the Kuala Lumpur International Airport (KLIA) (26km). The major industrial estates in Nilai are Nilai Industrial Estate, Arab Malaysian Industrial Estate, Nilai Utama Industrial Park, and Nilai Industrial Park [16]. These industries include electronic and electrical, textiles, food, paints, packaging and machinery manufacturing. Therefore, it is expected that particles released from the road and industries activities affecting the people who exposed to the increase of air pollution in Nilai. The site located near to railway stations operated by Keretapi Tanah Melayu Berhad (KTMB) in the northeast of Nilai town, close to main roads and influenced by emissions of industrial activities by a paint manufacturer to the northwest (within 500m); quarries activities to the northeast (1km) and a concrete ready-mix facility to the east (within 500m).

Sub-urban site located in Seberang Jaya, Pulau Pinang which is north-west coast of Peninsular Malaysia. This monitoring site located in the residential area near a primary school. It is also very near to Perai Industrial area (3km), the main industrial area in Pulau Pinang. The main economic activities that governed Seberang Jaya are services commercial and business area such as shopping mall (500m), hospital (500m), and a few accommodation facilities i.e. hotels (500m). Seberang Jaya is located in the vicinity of heavy traffic area as it is near to the North-South Expressway (300m) and Butterworth – Kulim Expressway (1km). Besides, the site is surrounded by residential areas and educational facilities i.e. government school which make Seberang Jaya experienced rapid development by growing populations.

## 2.2 PM<sub>2.5</sub> Concentration Sampling

PM<sub>2.5</sub> data collection has been started from 28<sup>th</sup> December 2014 until 22<sup>th</sup> May 2016 which covered for both sites. PM<sub>2.5</sub> concentration and meteorological parameters (wind speed and wind direction) were measured continuously by using Met One Instrument (E-BAM), a portable real time sampler that meet the USEPA requirement of Federal Equivalence Method (FEM) for Class III PM<sub>2.5</sub> measurement (40 CFR Part 53) [21] with PM<sub>2.5</sub> cut size inlet. Sequences 1h tape advanced was set on E-BAM to collect PM<sub>2.5</sub> sample by using glass fibre filter tape and conducted for 12 h per day for each site starting from 7 a.m. to 6 p.m. during daytime only. Total hourly samples collected for all site is 168 filter samples (n = 168) as shown in Table 2.

**Table 2 - Details on number of samples collected at each site**

Category	Hourly samples no.	Total hourly samples	Total hourly samples per site
Industrial	7 days x 12 hourly samples/day	84	84
Sub-urban	7 days x 12 hourly samples/day	84	84
Total samples			168

## 2.3 Wind Rose Plot and HYSPLIT

A wind rose predominantly represents the circulation and distribution of local wind patterns. Wind rose may give detailed information on how wind speed and direction are typically distributed at a particular location. Wind rose shows wind blowing from a particular direction and it is presented in a polar format. Length of each 'spoke' around the circle represents speed of the wind blowing from a specific direction. There were sixteen cardinals of wind directions and wind speed was measured in meter per second. Back trajectory analyses were used to determine the potential sources of air mass from a different direction at industrial and sub-urban site. Hybrid Single Particle Lagrangian Intergrated Trajectory (HYSPLIT) model has been used to identify the PM source regions and transport height of the polluted air masses [22-23]. The Global Data Assimilation System (GDAS, 1 degree, global, 2006-present) from NOAA's Air Resources Laboratory was used as input data for the model. The trajectories were driven using gridded meteorological data at six-hour time intervals with calculations set for 30 m, 150 m and 500 m above ground level. The results were presented as the mean trajectories of the clusters obtained for each month (based on sampling date).

## 2.4 Diurnal Variation of Hourly PM<sub>2.5</sub> Concentrations

Variation of diurnal was observed by using twelve hourly averaged (7 a.m. to 6 p.m.) at each site. Based on the diurnal plot, hourly trends, time for minimum and maximum concentration were determined.

## 2.5 PM<sub>2.5</sub> Sample Collection

Particles are deposited throughout the depth of the fibrous filter matt rather than on the surface. Prior to weighing, the filters were stored under a constant temperature and humidity condition of 25 °C and 50%, respectively for 24 hours [24]. Under normal operating conditions, particles deposited from a sample flow at a rate of 16.7 L min<sup>-1</sup> are focused on an 11mm diameter spot and run continuously for 12 h between 7:00 a.m. and 6:00 p.m. for all sites. The tape advances were set for every one hour but when the pressure drop was detected by E-BAM due to excessive particle loading, the tape will be advanced automatically. In a laminar flow hood, the filtered tape was unrolled and the spot was deposited and removed by 13 mm diameter steel punch (Eksucces, Woodridge, Illinois, USA) and cleaned with 70% methanol (CH<sub>3</sub>OH)-soaked Kimwipes (Kimberly Clark, Dallas, Texas, USA) between punches. The unexposed distance between E-BAM deposit spots is 14mm. Hence, it will allow a punch of 13 mm unexposed tape to be taken from the area as blank values for subtraction. Ambient concentrations for each sampling spot were measured in µg m<sup>-3</sup>.

The exposed filters collected at the monitoring site were labelled and directly stored. A sample labelling system was created in order to ensure the identification and distinguished of samples easier. Each filter was punched and assigned with an ID code including time and date of sampling and stored in an individual Petri Dish (Steriplan, Soda, lime glass, 90 x 15 mm, Duran). The petri dish was then wrapped with parafilm (#PM996, Pechiney Plastic Packaging, Menasha, Wisconsin, USA), and stored in the freezer at -4°C. Upon the completion of sampling periods, the samples were transported in a cool box with desiccant agent to the Environmental Laboratory, School of Civil Engineering, Universiti Sains Malaysia, for analysis.

## 2.6 Gravimetric Analysis

The filters were allocated in a desiccator and exposed in humid conditioned at 20°C to 30°C and relative humidity of 30-40% for at least 24 h before and after sampling [21]. The filtered samples were weighed using electric micro-weighing balance (Sartorius model CP2P-F) with a minimum resolution of 0.001 mg at temperature range between 20°C and 25°C and relative humidity between 32% and 45%. Mass of filter tape for both conditions (pre and post weighed) sampling was obtained as the average of three measurements, with observed variations were less than 5%. The difference between pre and post weighed was considered as actual loading of PM<sub>2.5</sub> on the filter tape. The mass concentration  $C$  (mg m<sup>-3</sup>) of the loaded filter was obtained from the relation (Eq. 1):

$$c = \frac{w_f - w_i}{v} \quad (1)$$

where  $w_f$  and  $w_i$  are the final and initial average weights of the filter after and before sampling, respectively, and  $v$ , the volume of air sampled.

## 2.7 Chemical Preparation and Extraction Procedure

Method of digestion was based on inorganic compendium method (IO 3.1) [22]. A punch of 13 mm diameter from the filtered sample was divided into two equal portions using sterile disposable scalpel (N0146687, Size 10, Kiato, Hannover, Germany). One portion was used for elemental analysis using ICP-OES (Varian 715-ES) and remained portion was stored as a backup. Vinyl gloves or plastic forceps were used to retrieve the filters and placed in a labelled 150 mL Griffin beaker. The filtered sample was placed down into the lower portion of the beaker to ensure that acid volume will cover the entire sample.

A preset calibrated automatic dispensing pipette or a Class A glass pipette was used to add 10 mL of extracting acid (HNO<sub>3</sub> and HCl, 3:1 (v/v) each) for analysis. The beaker was placed on a hot plate provided in a fume hood and refluxed gently while covered with a watch glass for 30 min at 85 °C. The sample was not allowed to dry. Then, the beakers were removed from the hot plate and cooled for 30 mins. Afterward, the residue was shaken, filtered through a filter paper with 0.22 µm pore size (Millipore), diluted to 50 mL with deionized water, and stored in 50 mL polyethylene bottles. These solutions were kept in a refrigerator at 4 °C before analysis. The samples were subsequently analyzed for 13 elements, in order of reactivity by referring to IUPAC, namely, Na, Mg, Al, K, Ca, Ti, Cr, Mn, Fe, Ni, Zn, Sr, and Pb using ICP-OES (Varian 715E).

## 2.8 Quality Assurance and Quality Control (QA/QC)

For the QA/QC, all the glassware and polyethylene containers used for the elemental analysis was washed with acids (10% nitric acid v/v) for 24 hour [23] and were rinsed several times with deionized water before use [24]. In

addition, all element regression coefficients must be ensured better than 0.999 before the ICP sample analysis. To validate the digestion method efficiency, the Standard Reference Material SRM 1648a urban PM was extracted and analyzed by ICP-OES to simulate the loaded filter samples. Not less than 20.0 mg of urban particulate matter (NIST SRM 1648a) was added to a beaker. The SRM was then ready for extraction procedure similar to the sample digestion [22].

The performance of the analytical method to quantify the metal elements in particulate matter was performed according to the criteria established by International Conference on Harmonization [25] guidelines; specificity, accuracy, precision, linearity and limit of detection and limit of quantitation as shown in Table 3. For specificity, six blank filter samples were acquired, each of blank contained 50.0 ml of the standard solution and five of them with following volumes: 1.25 ml, 2.5 ml, 5 ml, 10 ml, and 20 ml of the SRM1648a standard solution of 400 mg/l [22] and ICP-OES is used for analysis of each element's concentration. Pearson's coefficients from the standard curve give results between 0.9994 and 0.9999, fulfil the recommendation by ICH [25] ( $r \geq 0.90$ ). In order to determine the accuracy, the measured concentration between certified values was compared. It was expressed in percentage recovery (%). Based on ICH [25], and approved by research quality-assurance plan, the satisfactory accuracy in all elements was needed in the range within 80 to 120%. It corresponded to the uncertainty of the NIST certified values. External calibration curve was made to evaluate the linearity with multi element standard solution with the following five standard concentrations: 10, 20, 40, 80, and 160 mg/l.

**Table 3 - Detection limits of the analysed chemical compounds determined from 10 blank filters, the recovery rates of the elements as obtained with the applied HNO<sub>3</sub>/HCl extraction method.**

Element	Wavelength (nm)	Coefficient (r)	Recovery (%)	LOD (ppm)	LOQ (ppm)
Na	588.995	0.9999	106	9.49	31.64
Mg	279.553	0.9999	102	7.40	24.69
Al	396.152	0.9999	103	1.89	6.31
K	766.491	0.9994	113	1.41	4.72
Ca	396.847	0.9999	102	8.18	27.26
Ti	334.941	0.9999	96	0.43	1.43
Cr	283.563	0.9997	96	2.73	9.12
Mn	257.610	0.9999	101	0.32	1.08
Fe	238.204	0.9998	99	2.57	8.56
Ni	230.299	0.9997	95	0.06	0.21
Zn	213.857	0.9999	109	1.32	4.41
Sr	407.771	0.9999	94	7.29	24.30
Pb	220.353	0.9998	105	0.26	0.88

\*The chemical elements are arranged in accordance with the IUPAC; LOD- limit of detection; LOQ- limit of quantification

A limit of detection (LOD) and a limit of quantification (LOQ) for each element were calculated from the standard deviation (SD) of 10 blank filters. The LOD was calculated for three times SD and the LOQ for 10 times SD [25]. Standard deviations of 10 blank filters were used to calculate the limit of detection (LOD) and a limit of quantification (LOQ) for each element. ICH [25] refers to the 3:1 signal-to-noise ratio for LOD, while LOQ has been described as the concentration that provides a signal-to-noise ratio (peaks with a height of at least 10 times higher as the baseline noise level) of 10: 1.

In order to determine the concentrations of the elements, the elemental analysis was performed using ICP-OES (Varian 715-ES). Its high specificity, multi-element capability and good detection limits result in the use of the technique in a large variety of applications [26]. This instrument uses superheated argon plasma of 7000–10000 K to breakdown and excites the atoms of different elements to a higher energy level and then identifies the elements from the characteristic wavelength emitted during the excitation process. Table 4 shows the instrument setting of ICP-OES.

## 2.9 Source Apportionment of PM<sub>2.5</sub> Trace Elements using Principal Component Analysis (PCA)

PCA had been used to determine contribution of chemical components as well as to group the variables in order to determine the possible sources. Hourly averaged of PM<sub>2.5</sub> concentration and trace elements that resulted from the ICP-OES analysis was used in the analysis by using IBM Statistical Package for the Social Sciences (SPSS Version 22.0, USA). PCA was performed to reduce the data and extract a small number of factor (principal components, PCs) which would explain the variance observed in a larger number of variables in a more effective and efficient way [27].

**Table 4 - Instrument settings for the ICP-OES**

Instrument Setting	
RF power	1000 W
Nebulizer	Ultrasonic with membrane desolvator
Sample cone	Nickel, 1.1 mm orifice
Skimmer cone	Nickel, 0.9 mm orifice
Argon flow rates	
Plasma gas	15.0 L/min
Auxiliary gas	1.5 L/min
Nebulizer gas	1.17 L/min

Firstly, quality control measures were applied using Kaiser-Mayer-Olkin Measure ( $KMO \geq 0.5$ ) and Bartlett’s Test of Sphericity ( $p < 0.05$ ) [27] to assess the sufficiency of monitoring data for PCA. According to Chattopadhyay and Chattopadhyay [28], KMO role as an indicator whether PCA is suitable for removing multicollinearity in the monitoring data or not. The correlation pattern is relatively compact and suitable for PCA if the KMO value close to 1, however when the KMO value is close to 0, it indicates that the data is not suitable for PCA. The equation for KMO measures is given in (Eq. 2):

$$KMO = \frac{\sum_{i \neq j} \sum_{i \neq j} r_{ij}^2}{\sum_{i \neq j} \sum_{i \neq j} r_{ij}^2 + \sum_{i \neq j} \sum_{i \neq j} a_{ij}^2} \tag{2}$$

where,  $r_{ij}$  = simple correlation coefficient,  $a_{ij}$  = partial correlation coefficient between  $i$  and  $j$ . In PCA, KMO and Bartlett’s test of sphericity ( $p < 0.001$ ) normally combined in order to determine the applicability of the monitoring records for PCA analysis [29]. Eq. (3) expressed Bartlett’s test of sphericity measure.

$$x^2 = -\left(n-1-\frac{2p+5}{6}\right)l_n|R| \tag{3}$$

$$H_0 : |R|=1, H_1 : |R| \neq 1$$

where,  $n$ : number of observations,  $p$ : number of variables and  $R$ : correlation matrix.

In most cases, only total variance higher than 60% is considered significant [30]. According to Pires et al. [31], PCA creates new variables into several principal components (PCs) that are uncorrelated to each other in ascending order where the first PC (PC1) explains most of the variance of the original data variables. The second PC (PC2) explains lesser of the variance of the original data variables and so forth. PCs can be generalised as in Eq. (4) [32].

$$PC_i = l_{i1}X_1 + l_{i2}X_2 + \dots + l_{im}X_m \tag{4}$$

where,  $PC_i$ : principal component for  $i$ th,  $X_m$ : observed variable and  $l_{mi}$ : loading of the observed variable,  $X_m$ .

Factors were extracted with eigenvalues greater or equal to 1 ( $\geq 1$ ) in order to create the PCs group [33]. Eigenvalues were obtained by using Eq. (5).

$$|c - \lambda I| = 0 \tag{5}$$

where,  $c$ : correlation of the monitoring record,  $\lambda$ : eigenvalue and  $I$ : identity matrix. Varimax rotation technique was used in computation of factor rotation to clarify the underlying component and to find attributed contribution that influenced each of the variables [31, 33, 34].

### 3. Results and Discussion

#### 3.1 Variation of PM<sub>2.5</sub> Concentration

PM<sub>2.5</sub> concentrations has been compared with the permissible international standards, which are USEPA, WHO and MAAQS (IT-2) in 2018 in order to know concentration level of PM<sub>2.5</sub>. Fig. 2 shows the boxplot for PM<sub>2.5</sub> concentrations at industrial and sub-urban sites. The mean concentration of PM<sub>2.5</sub> recorded in industrial site was 46.68  $\mu\text{g m}^{-3}$ , almost 2 times higher than WHO limit (25  $\mu\text{g m}^{-3}$ ) and more than 1 times higher than USEPA limit (35  $\mu\text{g m}^{-3}$ ). However, mean concentration of PM<sub>2.5</sub> at sub-urban site (16.67  $\mu\text{g m}^{-3}$ ) did not exceed all standards and it is still under the maximum limit as suggested by MAAQS IT – 2 (50  $\mu\text{g m}^{-3}$ ). However, mean concentrations get in this study

were calculated based on 12 hr averaged not 24 hr averaged that has been used in standards. The highest mean at industrial site was found to be higher compared with Balasubramanian et al. [35] in Singapore ( $27.2 \mu\text{g m}^{-3}$ ), Mohd Tahir et al. [11] at the East coast of Peninsular Malaysia ( $14.3 \mu\text{g m}^{-3}$ ) and recently by Ling et al. [10] ( $30 \pm 7 \mu\text{g m}^{-3}$ ) at Kuala Lumpur.

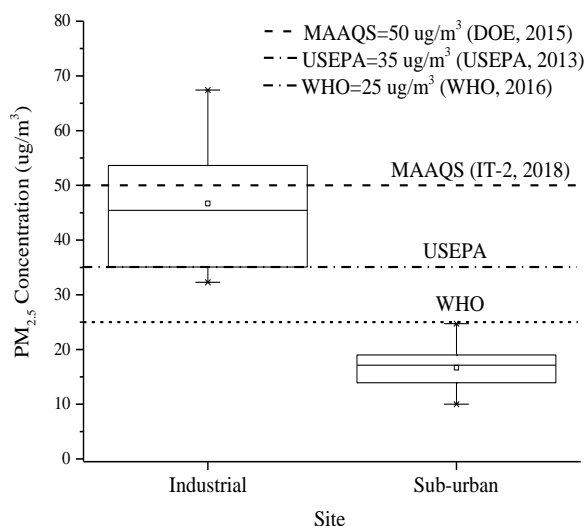


Fig. 2 - Boxplot for  $\text{PM}_{2.5}$  concentration monitored at both sites

### 3.2 Site Profiling of Possible Sources based on Wind Rose and HYSPLIT

The distribution of possible sources including industrial, construction and open burning activities surrounding site area during the monitoring is classified by referring the wind distribution by the wind rose plot for each site. Typical changes in wind direction occurred every day in the site.

At industrial site, dominant wind direction was from south-east with the highest wind speed recorded was  $2.01 \text{ ms}^{-1}$ . The dispersion of  $\text{PM}_{2.5}$  was influenced by the prevailing winds, which might transported  $\text{PM}_{2.5}$  from nearby sources (industrial and construction activities). Textile industries and construction for the development purpose of commercial areas (i.e. double storey shop) are among the contributors of  $\text{PM}_{2.5}$ . At construction site, usually during the construction phase, there are uncovered surfaces. This condition might cause the dust uplift during the movement of vehicles going in and out from the site. High wind speed with  $1.8 \text{ m s}^{-1}$  was recorded that blew from east-northeast where there were industries activities at that area. The quarry is located about within 500 meter from the monitoring station. Madungwe and Mukonzvi [36] stated that the extraction of minerals from surface mines and quarries such as blasting, road haulage, loading, crushing and stockpiling produced significant fugitive dust emissions as a result of site activities which contribute to the ambient  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  concentrations. Dust generated from surface mining sites are the result of a force applied to bulk material such as rock for economical extraction, handling, processing, storage and transportation [37].

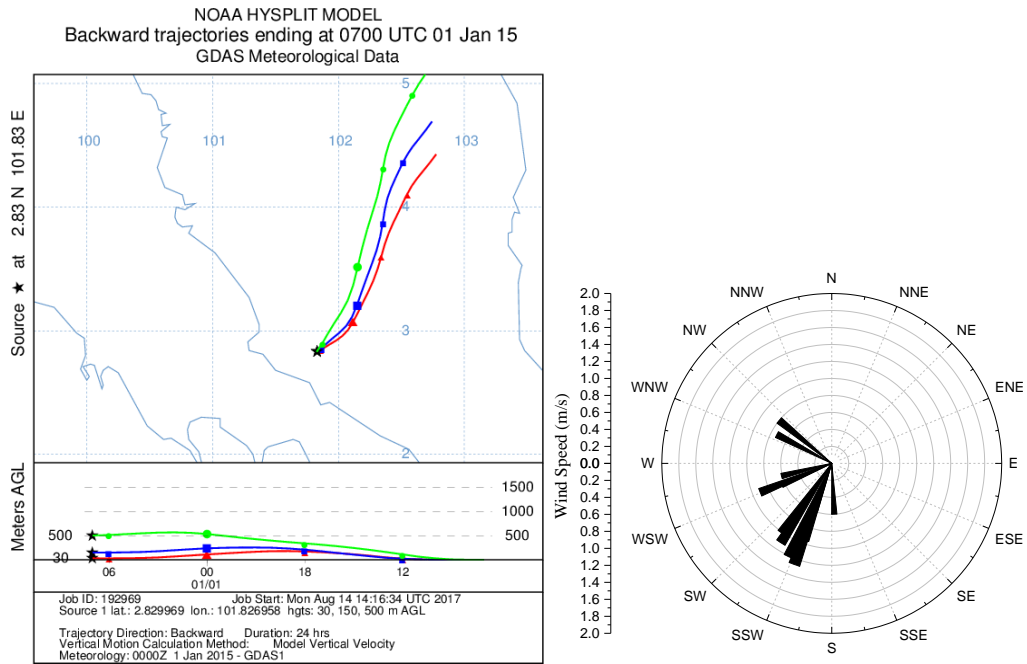
HYSPLIT backward trajectories and wind rose analysis for industrial site is illustrated as in Fig. 3. The figure demonstrates that during the study period, the winds mainly blowing from the north east direction where the southern China Sea is located. The differences of wind variation occurred because of the wind data recorded using E-BAM was taken at a certain level that less than 1.5 m from the ground level. The readings are known as local wind direction had been influenced by the topography and the location of the sampling sites.

Dominant wind direction at sub-urban was from the southwest direction (blowing from southwest to northeast) with the highest wind speed of  $0.70 \text{ m s}^{-1}$ . HYSPLIT backward trajectories and wind rose analysis for sub-urban is depicted in Fig. 4. The winds mainly blowing from the south east direction. It can be seen that, at this direction, there were few industrial sources that located about 1-2 km away from the monitoring site. Besides industrial activities, traffic density, population density, and different transportation system were available around the site that might contributed to high  $\text{PM}_{2.5}$  concentration. Sub-urban site is a central for Seberang Perai which has distinct employment bases that make it a busy network during peak hour.

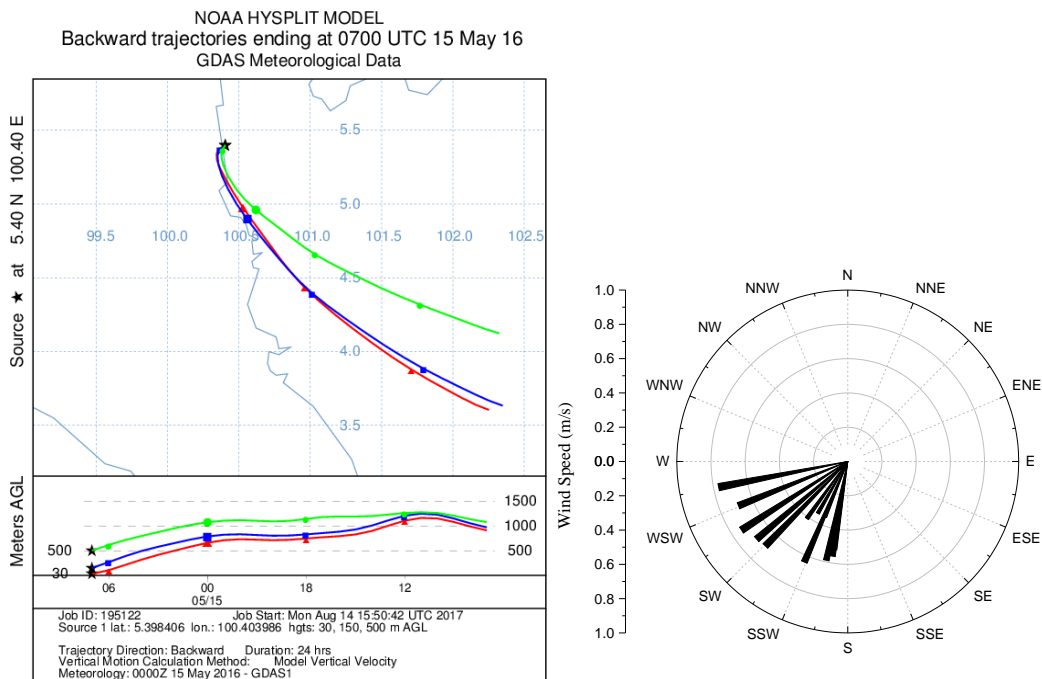
### 3.3 Diurnal Variations of $\text{PM}_{2.5}$ Concentration at Both Sites

Diurnal variations of hourly  $\text{PM}_{2.5}$  concentrations is depicted in Fig. 5. Diurnal variations of  $\text{PM}_{2.5}$  concentration were observed at both sites, but at different hourly patterns. Meteorological conditions, particularly atmospheric stability and wind speed can cause late evening peak [38-39]. Industrial site clearly recorded the highest concentration. This phenomenon is considered to be correlated directly with the location of site which nearby the industrial area, roads

as people are going to work and caused the uplifting of particles. The lowest values of PM<sub>2.5</sub> concentration occurred in the morning (11:00 a.m.) might be due to less influence of anthropogenic activities on fine particulate levels during that time.



**Fig. 3 - HYSPLIT backward trajectory (7 days) analysis for air masses and wind rose plot (blowing from) (12hr) at industrial site**



**Fig. 4 - HYSPLIT backward trajectory (7 days) analysis for air masses and wind rose plot (blowing from) (12hr) at sub-urban site**

PM<sub>2.5</sub> concentration at sub-urban site showed a distinctive diurnal pattern while the lowest values observed during day time (11:00 a.m.). However, in general, the wind speed increases substantially during the day had caused dispersion of aerosol particle mass. PM<sub>2.5</sub> exhibited a bimodal pattern with two marked peaks at industrial site, during morning rush hours (08:00 hrs –09:00 hrs) and evening rush hours (15:00 hrs). Similar observations and implications



were reported by Chan and Kwok [40]. Meanwhile, at sub-urban site, the peak occurred in the evening at 14:00 hrs and 17:00 hrs.

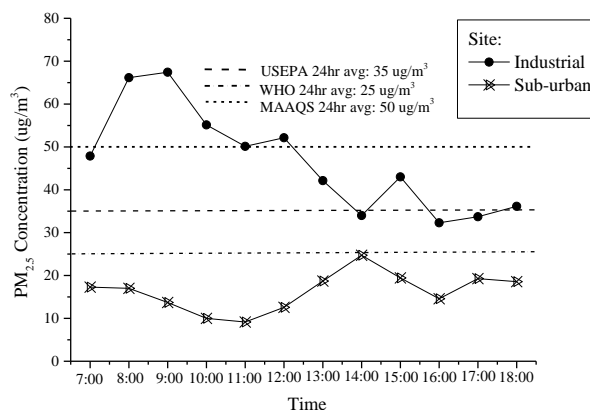


Fig. 5 - Diurnal variation of hourly PM<sub>2.5</sub> concentration (12 h) at both sites

### 3.4 Variations of PM<sub>2.5</sub> Trace Elements

The average concentrations and standard deviation of the detected elements measured in both sites are summarized in Table 5. Differences in average of analyzed elements indicated that concentrations were different, as it was influenced by locations and sources of trace elements at site.

The levels of PM<sub>2.5</sub> chemical concentration in industrial and sub-urban sites shown in Fig. 6 exhibited different patterns because of the different source origins. Al, K, Ca and Fe were major chemical components of PM<sub>2.5</sub>; ranged from  $451 \pm 387 \text{ ng m}^{-3}$  to  $1472 \pm 1201 \text{ ng m}^{-3}$  at industrial site. The concentrations for other components, including Cr, K, Mg, Mn, Na, Ni, Pb, Sr, Ti, and Zn, were in the range of  $0.2 \pm 0.1 \text{ ng m}^{-3}$  to  $49 \pm 17 \text{ ng m}^{-3}$ . Al is the highest concentrations in both sites. Ca and Fe dominated in industrial site compared to sub-urban which dominated by Na and Al. Al, K, Ca, and Fe elements are indicators of crustal weathering and mineral dust [10], [12], [40], [41]. High concentration of Al and Ca most likely originated from resuspension of road dust while high concentration of Fe can be attributed to traffic emissions and soil dust [10]. According to Caggiano et al. [43], high concentrations of Fe, particularly in PM<sub>2.5</sub>, are associated with exhaust emissions, especially from gasoline and diesel-fuelled road vehicles. Several studies have also found that soil dust is a major source of Fe [10], [12], [42].

In sub-urban site, concentrations Na, Al, K, and Zn elements were higher than other trace elements with concentrations  $190 \pm 44.5 \text{ ng m}^{-3}$ ,  $526 \pm 131 \text{ ng m}^{-3}$ ,  $98 \pm 67 \text{ ng m}^{-3}$  and  $67 \pm 18.5 \text{ ng m}^{-3}$ , respectively. Al was detected as the dominant element in sub-urban samples compared to others element. High Na, Al and K concentrations observed at sub-urban site are caused by crustal element as found by Gugamsetty et al. [44]. Zn is widely used as a chemical fingerprint for tyres wear especially those with two strokes engines, such as motorcycles and motor scooters [44] and also as a tracer for unleaded fuel and diesel-powered motor vehicle emissions.

Table 5 - Concentrations of the elements identified in PM<sub>2.5</sub> samples (12 h mean  $\pm$  sd) (ng m<sup>-3</sup>)

Element	Industrial	Sub-urban
N	84	84
Sodium (Na)	$49 \pm 17$	$190 \pm 44.5$
Magnesium (Mg)	$36 \pm 31$	$5.5 \pm 4.5$
Aluminium (Al)	$1472 \pm 1201$	$526 \pm 131$
Potassium (K)	$567 \pm 412$	$98 \pm 67$
Calcium (Ca)	$858 \pm 848$	$30 \pm 6$
Titanium (Ti)	$45 \pm 38$	$0.5 \pm 0.3$
Chromium (Cr)	$39 \pm 15$	$0.5 \pm 0.3$
Manganese (Mn)	$0.9 \pm 0.5$	not detected
Ferum (Fe)	$451 \pm 387$	$3 \pm 2.3$
Nickel (Ni)	$0.2 \pm 0.1$	$0.1 \pm 0.04$
Zinc (Zn)	$22 \pm 13$	$67 \pm 18.5$
Strontium (Sr)	$20 \pm 4$	$6.5 \pm 4$
Lead (Pb)	$14 \pm 13$	$0.44 \pm 0.32$

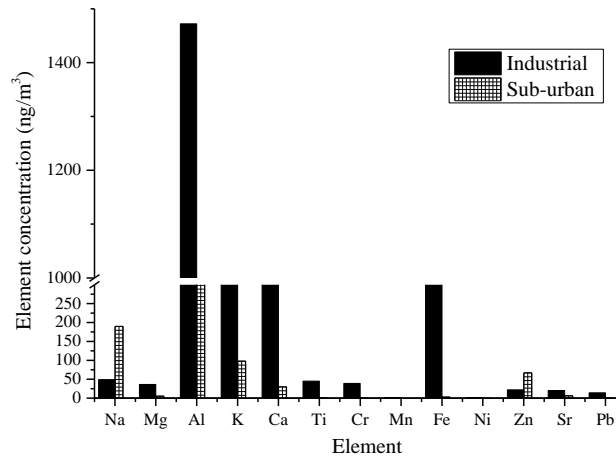


Fig. 6 - Average elemental concentrations in industrial and sub-urban site

### 3.5 Source Apportionment - Principal Component Analysis (PCA)

PCA had been applied to the trace elements and results of factor loading after varimax rotation for all sites are shown in Table 6. Validation of PCA results was based on Bartlett’s test of sphericity and Kaiser-Meyer-Olkin (KMO). Results from Bartlett’s test show test statistics value  $p < 0.001$  which suggested that selected variables were significantly related to each other. Meanwhile, KMO value recorded from all types of the area show that the values exceeded 0.5. Hence, both sites were suitable to perform PCA. Three factors with various sources of  $PM_{2.5}$  were obtained for both sites. Factor loading  $\geq 0.40$  are considered in this results, but there are few elements (Mg, Cr, Mn, and Ni) were not listed in the PC because the factor loading  $\geq 0.4$ . To overcome this, factor loading  $\geq 0.30$  was taken into account for that element at sub-urban site. The total variations of PCs were 81.45% and 69.79% for industrial and sub-urban, respectively.

Table 6 - Results of principal component analysis after varimax rotation at both site

Site	Accepted if;	Industrial			Sub-urban			
KMO	$\geq 0.5$	0.891			0.826			
Bartlett’s Test	$p < 0.05$	$< 0.001$			$< 0.001$			
PC		PC1	PC2	PC3	PC1	PC2	PC3	
Element		C	I	V	C	I	V	
Na	FL $\geq 0.4$	0.85	-	-	0.83	-	-	
Mg		0.54	-	-	0.68	-	-	
Al		0.79	-	-	0.91	-	-	
K		0.79	-	-	0.84	-	-	
Ca		0.92	-	-	-	0.82	-	
Ti		0.88	-	-	-	0.84	-	
Cr		0.83	-	-	-	-	-	
Mn		-	0.90	-	-	-	-	
Fe		-	0.91	-	-	-	-	0.87
Ni		-	0.93	-	-	-	-	-
Zn		0.82	-	-	-	0.86	-	-
Sr		0.86	-	-	-	0.91	-	-
Pb		-	-	0.72	-	-	-	-0.44
Eigenvalue		$\geq 1$	7.77	1.82	1.00	4.63	1.58	1.04
V (%)		59.77	13.98	7.70	43.63	15.74	10.42	
$\Sigma V$ (%)	$\geq 60\%$	58.77	73.75	81.45	43.63	59.37	69.79	

Note: KMO: Kaiser-Meyer-Olkin; PC: Principal Component; C: Crustal; I: Industrial; V: Vehicular; V is variance;  $\Sigma V$  is total variance explained; FL is factor loading

For industrial site, PC1 represents 59.77% of total variations of the data set and dominated by Na, Mg, Al, K, Ca, Ti, Cr, Zn and Sr. Other researchers reported that these elements as markers for a crustal material [6, 10, 12, 40, 42, 43, 45]. Given that the sampling site was located near a main road, vehicular movement might cause suspensions of soil/road dust. This is also observed by Ny and Lee [14]. PC2 explained 13.98% of the total variance, with high loadings of Mn, Fe, and Ni indicate that source was coming from the industrial activities. Vehicle exhaust can contain

Cr, Fe, Zn and Ni [12, 41]. Given that the sampling site was near industrial area, prevailing winds could play a significant role in spreading the elements to the monitoring area. Fe can originate from the welding activities around the industrial area in that site, and Mn might exist in natural and anthropogenic sources. Ni is widely reported in the literature as markers for the residual oil combustion [42-44]. PC3 was from vehicular sources dominated by Pb which explained 7.70% of the total variance. Pb in industrial area is emitted by different sources and each of these industries emits Pb in different chemical form. Begum et al. [46] found that Pb in  $PM_{2.5}$  was released from the pre-existing road dust.

For sub-urban site, PC1 represents 43.63% of total variations of data set and given that it presents high factor loadings for Na, Mg, Al, K, Cr, Ni, Zn, and Sr, common elements that were grouped as crustal material in industrial and urban sites. In sub-urban, fewer elements have high factor loading compared to industrial and urban. The location of sampling site was surrounded by residential area and schools, might influenced the element concentrations. PC2 explained 15.74% of total variance, with high loadings of Ca, Ti and Mn, was from industrial sources. Mn most likely originated from industrial activities especially in domestic manufacturing and processing facilities. Ca, Ti and Mn also can be found from resuspended road dust. A previous study reported a strong contribution of road dust by vehicles movements on road surface to  $PM_{2.5}$  [6, 12]. PC3 was from vehicular sources dominated by Fe and Pb which explained 10.42% of the total variance. Vehicle exhaust can contain Fe, as found by Srivastava et al. [41] and Cheng et al. [42] in diesel-fuelled vehicles.

#### 4. Conclusion

This research was embarked upon motivation to investigate  $PM_{2.5}$  concentration, trace elements, and source apportionment of  $PM_{2.5}$  of two categories of monitoring sites, namely industrial and sub-urban site. The highest mean of  $PM_{2.5}$  concentration was recorded at industrial site ( $46.68 \mu\text{g m}^{-3}$ ) and the lowest mean of  $PM_{2.5}$  concentration was recorded at sub-urban site ( $16.67 \mu\text{g m}^{-3}$ ). Results on trace elements analysis revealed that both sites were dominated by Al. Mean concentrations of Fe element in  $PM_{2.5}$  were found to be the highest among other elements. The total variations produced could explain the numbers of PCs in the data were 81.45%, and 69.79% for industrial and sub-urban, respectively. Three principle components for industrial site are identified, which are PC1 (crustal: Na, Mg, Al, K, Ca, Ti, Cr, Zn, and Sr), PC2 (industrial: Mn, Fe, and Ni), and PC3 (vehicular: Pb). The numbers of PCs indicate the group of possible sources, with crustal material was found dominant at each sites. For sub-urban site, PC1 (crustal: Na, Mg, Al, K, Cr, Ni, Zn and Sr), PC2 (industrial: Ca, Ti and Mn), and PC3 (vehicular: Fe and Pb). Industrial site has different group characterization compared to sub-urban. In conclusion, higher  $PM_{2.5}$  concentrations are recorded in location with high anthropogenic activities (e.g. industry emission, vehicles emission). The results obtained from this study proved that  $PM_{2.5}$  concentration and trace elements depend on the location of sampling area, meteorological factors and types of local sources. Hence, from this study, it may help local authorities to observe and predict the high concentration of  $PM_{2.5}$  in problematic areas.

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