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ACS Earth Space Chem., **Just Accepted Manuscript** • DOI: 10.1021/
acsearthspacechem.0c00103 • Publication Date (Web): 14 Jul 2020

Downloaded from pubs.acs.org on July 14, 2020

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**Characteristics, emission sources and risk factors of heavy metals in PM_{2.5} from
Southern Malaysia**

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Abstract

Exposure to fine particulate bound toxic metals in ambient air poses adverse effects to human. This study aims to determine the spatial variability in heavy metals in PM_{2.5} samples, for identifying their potential sources and to perform the health risk modelling. PM_{2.5} samples were collected using high volume sampler (HVS) on 24 h basis from three sites in Johor areas in Malaysia from January to March 2019. Metals were initially extracted using microwave assisted digestion and the metals concentrations were analysed using inductively coupled plasma mass spectroscopy (ICPMS). Overall, the abundant metals in PM_{2.5} among the metals analyzed were Zn with mean (29.92 ng/m³) and Se with mean (27.02 ng/m³). The sources of PM-bound metals were identified using absolute principal component score (APCS) with multiple linear regression (MLR). The major source contribution was noted from vehicle emission (41%). Other potential sources for the metals in PM_{2.5} was from oil coal fired power plant (34%) and oil refinery and industrial emission (4%) leaving 22% of metals undefined. From the health risk analysis, the hazard quotient (HQ) and excess lifetime cancer risk (ELCR) values of the metals were within the tolerance level. The trend for HQ values were Co < Zn < Pb < Cu < Ni < As for adolescent and Co < Zn < Cu < Pb < Ni < As for adult age. Whereas for ELCR values, the trends were same for both adolescent and adult age groups as Pb < Ni < As. Few of the toxic metals showed comparatively high HQ values that might be a risk in the long-term exposure. Considering the highest noted contribution from vehicular emissions, it is advised to raise public awareness to practice carpooling and use public transportation to reduce emissions from vehicular sources.

Keywords: Fine particulate matter; Trace metals; Absolute principal component score; Hazard quotient; Carcinogens

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Highlights

- Zn and Se were predominant among the metals analysed in PM_{2.5}
- Local meteorology impacts potentially on the heavy metals in PM_{2.5}
- Vehicle and coal-fired power plant are dominant sources of PM_{2.5}-bound metals
- Adolescent are vulnerable to non-carcinogenic risk

1.0 Introduction

In recent decades, air pollution has been linked with many adverse human and environmental impacts. Among many criteria air pollutants, airborne fine particulates are one of the most

studied pollutants as it induces many negative environmental impacts like on ambient air quality, on visibility, on human health and on climate change. There are many documentary evidences that exposure to airborne fine particulates has resulted into adverse health impacts like respiratory and cardiovascular diseases and in many cases premature mortality¹⁻⁴. Very recently, World Health Organization⁵ recognized urban outdoor air pollutants and indoor air pollutants, especially airborne particulates as a major public health concern with more than two million premature deaths per year⁵⁻⁷.

Several studies reported fine particulate matter (PM_{2.5}, aerodynamic size less than or equal to 2.5 µm) pose deep concern as these tiny particle can penetrate deeper into the lungs alveoli upon inhalation⁸⁻¹³. Deeper penetration into the lungs causes PM_{2.5} to enter the bloodstream, which results in hypertension and disrupting the blood vessels and the damage may spread to the heart, causing damage to its cell structure and function^{14, 15} and can cause also to brain damage or responsible of neurological diseases e.g. Alzheimer's disease^{2, 16}.

PM_{2.5} contains a large variety of harmful elements; the impactful ones are particularly toxic heavy metal elements that may damage the human body. Toxic heavy metals cause harm to the human body by three main ways of exposure: food consumption, contact with the skin and inhalation^{17, 18}. Heavy metals in PM_{2.5} are considered to be the major toxic components. Biological damages were found induced in laboratory cells, animals and cohort population due to some metals associated with PM_{2.5}¹⁹. Air pollutants as well as PM_{2.5} impact on human health via deposition into the respiratory system were well documented also in Malaysia²⁰⁻²³.

It's essential to identify the sources of particulate matter as the kind of particulate sources like combustion, crustal sources, and marine sources, primarily regulate the fundamental properties of PM_{2.5} like its size, morphology, composition and thereby, showing implications to the receptor sites. Recent studies have reported variability in PM_{2.5} sources across Malaysia

therefore, further research is needed to detect and quantify the sources of fine particulate matter so that an effective air quality management plan can be implemented²⁴⁻²⁷. Among many available techniques, receptor models are widely used and convenient methods to determine the sources of PM_{2.5}²⁸. Therefore, to detect the sources of fine particulate matter, based on its composition, here we have used absolute principal component score (APCS) and multiple regression analysis (MLR) receptor models. APCS as a corrected version of principal component analysis (PCA) model was convenient to use as it provide fast identification of source and did not require any specific software to be used^{4, 28, 29}. APCS applies Z-scores proposed by Thurston and Spengler⁴ using a fictitious zero sample which appropriately apportion the sources of airborne compositions quantitatively without any source profile^{30, 31}. This work analyses the three months measurements of fine particulate matter and particulate-bound metal compositions collected at various representative locations in Johor state. In conjunction with the harms to human health and other related environmental issues of PM_{2.5}, this study focuses on determining the presence of heavy metals in PM_{2.5} samples, to identify the potential sources of heavy metals in PM_{2.5} samples in selected areas in Johor state and finally to establish the health risk of particulate-bound toxic heavy metals. Potential implications of this study will be to strengthen air quality management plan of the city and to understand source specific impacts of particulate matter to human health.

2.0 Methodologies

2.1 Details of the study areas

Stepping into 2020, Malaysia now is forging ahead in many industrial fields and the population of the nation keeps increasing (32,157,114 as of 31st of December 2019; Worldometers³²) equivalents to 0.42% of the world population. Due to rapid population growth and rapid

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3 industrialization, air quality in Malaysia becomes a major concern. Factories, power plants,
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5 vehicles, biomass burning are prominent sources that contribute primarily to air pollution. In
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7 this study, Johor state is chosen because the state is rich in economy, especially in the secondary
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9 and tertiary sectors (manufacturing and service sector). For this study, two cities in Johor
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11 namely Johor Bahru and Batu Pahat are selected for particulate sampling. Johor Bahru is the
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13 5th highly populated city in the country with a total population of 802,489 whereas the total
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15 population in Batu Pahat is 156,236 ³². Samples collected are from chosen educational
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17 institutions in Johor, which are Pusat Pembangunan Tenaga Industri Johor (PUSPATRI) in
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19 Pasir Gudang, Universiti Tun Hussein Onn Malaysia (UTHM) in Batu Pahat and Sekolah
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21 Menengah Kebangsaan Gelang Patah (SMKGP) in Gelang Patah. PUSPATRI and SMKGP are
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23 located in Johor Bahru city. Their geographical factor is one of the reasons these educational
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25 institutions are chosen. UTHM is located nearby Parit Raja industrial area, PUSPATRI is
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27 located nearby Pasir Gudang industrial area and SMKGP is located in a quite complex urban
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29 area but not too close to industrial area. Being located in the industrial region and complex
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31 urban zone, these sampling locations are in major risk for high contamination of airborne
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33 particulate matter in the atmosphere. A detailed description of the sampling locations has been
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35 provided in Table S1. Figure 1 shows the location of the three sampling sites in the state of
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37 Johor in Malaysia whereas topography of the region is included in Figure S1.
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45 2.2 Sampling procedures of PM_{2.5}

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48 Samples were collected every week from January to March 2019 from each of the monitoring
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50 stations. Particulate monitoring was performed by a high-volume air sampler (HVS) (Tisch,
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52 USA) using a quartz fibre filter (Whatmann, QMA, UK). A high-volume air sampler was used
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54 to collect airborne particles. The sampling time was at 1200 am for every station and was
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56 operated for a duration of 24 h basis each time. A total of eighteen samples were collected from
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January to March 2019 for each sampling stations. Table S2 shows the chosen samples for every sampling station and their respective filter ID and sampling flow rates.

2.3 Microwave assisted digestion and analysis of the trace metal compositions

Eighteen selected samples and eight blank filters were each cut into small pieces (3cm × 3cm). For each filter paper the area of the whole region which the air passed through was jotted down. Six reagent blanks were also prepared. The air particle samples were digested by wet acid digestion method, assisted by the microwave digester (MARS 6™, CEM, North Carolina, USA). The reagents used for wet digestion was aqua regia, HCl and HNO₃ with a ratio of 3:1. Filter paper was cut into smaller fragments into each Teflon XPRESS vessel. In each Teflon vessel, 6 ml of HCl and 2 ml of HNO₃ were added and then the mixture was let sit for 15 min in the fume hood cupboard before the vessels were capped and the lids were tightened. The programme used for the sample digestion was 500 W of power, ramping time of 45 min to 180 °C and hold time of 15 min. When the digestion was complete, the microwave was let cool until the temperature fell below 70 °C before the carousel of vessels in the microwave could be safely taken out. The lid of the vessel must be carefully opened and the process must be done in the fume hood cupboard as gases were released as the end products of the reaction. The solution was filtered gravitationally into 25 ml plastic volumetric flasks, using plastic filter funnels and Whatman glass fibre filter papers. Ultrapure water was used to dilute the solution up to the marks. Then, the sample and blank solutions were transferred into plastic 50ml vials, labelled and stored in the refrigerator prior to analysis.

The analysis was conducted using ICP-MS (model 7500, Agilent, USA). On the day of the analysis, standard solutions were freshly prepared from the multi-element ICP-MS calibration standard stock solution (Inorganic ventures, USA). Standard solutions were prepared in the range of 10 to 100 ppb. The standard solutions were diluted using the same method done in

method validation for QA/QC. Then, the standard solutions and the CRM solutions were run through ICP-MS for the heavy metal determination. Ten heavy metals (V, Cr, Ni, Cu, Zn, As, Se, Cd, Pb and Co) were analysed to determine their concentrations.

Table S3 shows the area of filter paper of each selected samples for heavy metals detection. From the results obtained by ICP-MS, the concentration of every heavy metals was corrected with the average blank filter paper concentration. To take into account, the concentration of heavy metals must be calculated for the whole filter paper, using the area of filter paper as shown Table S3.

2.4 Quality assurance and quality control (QA/QC)

As for QA/QC, all the plasticwares used for the elemental analysis were washed in acid bath mixture (2% nitric acid v/v) for 24 h and rinsed several times with deionised water and once with ultrapure water. Method validation was done by using environmental certified reference material (CRM) urban aerosol no. 28 which was developed and certified by the National Institute for Environmental Studies (NIES), Japan, purposely for the determination of multi-elements in aerosol particulate matter. The standard solutions for the ICP-MS run was prepared right before the analysis. The standard solutions and the solutions prepared using certified reference material (CRM) were run through ICP-MS for the heavy metal determination. Thirteen metals that were determined for CRM consisting of three high concentrations which were Mg, Ca and Fe meanwhile the low concentration metals were V, Cr, Ni, Cu, Zn, As, Se, Cd, Pb and Co. The CRM analysis was done separately for the low and high concentration metals. Prior to ICP-MS determination, the expected concentration of metals was calculated using the weight of CRM in Table S4, by taking the ratio from the actual concentration of metals as listed in the CRM certificate. From the results in Table S4, Table S5 and Table S6, the concentrations obtained were compared with the expected values and percentage recoveries

were calculated. As for the method validation by using aqua regia as the reagent for wet digestion, five metals which were Cr, Ni, Pb, Co and Fe showed good percentage recoveries with average value of 42% (Co), 97% (Fe), 56%⁹, 68% (Ni) and 69%.

2.5 Data analysis and chemometrics modelling

2.5.1 Statistical analysis

All statistical analysis in this study was performed using Microsoft® Excel 2010. Principal component analysis (PCA), absolute principal score (APCS) and multiple linear regression analysis (MLR) model were performed using Statistical Package for the Social Sciences (SPSS, Version 21.0, USA) and JMP Pro 15 (SAS, USA) software.

2.5.2 Receptor modelling

Principal component analysis (PCA) was applied to determine the sources of PM_{2.5}. Firstly, to know the sufficiency of monitoring data for PCA, quality control measures were applied using Kaiser-Mayer-Olkin Measure ($KMO \geq 0.5$) and Bartlett's Test of Sphericity ($p < 0.05$). KMO acts as an indicator in ensuring PCA is suitable for removing multicollinearity in the monitoring data. If KMO value is close to 1, the correlation pattern is suitable for PCA but vice versa if KMO value is close to 0²⁴. Our KMO results showed a value of >0.5 which suggested that the dataset was appropriate to conduct PCA analysis. However, a minimum but sufficient number of data set has been suggested by several researchers to obtain a statistically stable PCA results³³⁻³⁵.

By principle, PCA creates new variables into several principal components (PCs) which are uncorrelated to one another, where the first PC (PC1) will explain most of the variance of the original data variables whereas the second PC (PC2) will explain lesser of the original data variables. The list of the PCs is generalized as in the Eq. 1 below:

$$PC_i = l_{i1}X_1 + l_{i2}X_2 + \cdots + l_{im}X_m \quad (1)$$

Where PC_i is the principal component for i th, X_m is the observed variable and l_m is the loading of the observed variable. Due to the large uncertainty in the PCA derived factor scores and that followed by a multiple linear regression (MLR) to quantitatively apportion the sources, absolute principal factor scores (APCS) procedure was introduced by Thurston and Spengler⁴ to reduce the error in estimation of the sources. Thus, PCA coupled with APCS and MLR later widely has been applied to quantify the sources of $PM_{2.5}$ and other pollutants in air^{28, 29, 36}.

In the PCA-APCS-MLR, the data from three sites located in the Southern district of Malaysian Peninsula were combined as the input data of 18 samples. The three sites are located at the three educational institutes in the Johor Bahru. Two of them are suspected to receive emission of the pollutants from the industrial settings. However, one site is located in the vicinity of an urbanized setting. The geographical location of the three sites in Johor Bahru which passively receive emission of the heavy industrial activities from nearby Singapore, northeasterly Indochina region as well as localized pollutants. A comprehensive interpretation of the identified sources was illustrated for the transported and local sources using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) modeling. Thus, we presume that the nature of the source might be similar for the three sites and will represent the Johor region but the strength of the emission by the individual source possibly will vary to each site. Thus, the concentration data of the variables from the three sites were merged to run the PCA. The contribution of the identified sources estimated for the individual sites using the hybrid of APCS-MLR method.

2.5.3 US EPA Health risk modelling

In this study, the model used to estimate the exposure of students to heavy metals was also developed by the US EPA³⁷. US EPA models were widely applied by researchers in the health risk assessments. Health risk assessments was further divided into two parts, non-carcinogenic

and carcinogenic health risks as reported in the literature ^{25, 27}. Metals and metal compounds both have quite diverse toxicological profiles. For the purpose of risk assessment, critical effects served as the basis to derive the benchmark toxicity values. These effects are defined as the first adverse effect, also known as the precursor that occurs to most sensitive species as the dose rate of an agent increases ³⁸. IARC ³⁹ had classified at least five transition metals which are As, Cd, Cr, Be and Ni as carcinogens to humans in one form or another or in particular routes of exposure, Cd and Ni were classified as Class 1 carcinogenic elements while Pb as Class 2B carcinogenic element. As, Cd, Cr, Ni and Pb were recognized as carcinogenic metals and they were ingested by exposure via the inhalation pathway ⁴⁰. EPA's IRIS program had stated Pb compounds as probable human carcinogen. Whereas other metals, there are mixed evidences about their potential and carcinogenic risks.

2.5.3.1 Health risk assessment for non-carcinogenic metals by inhalation

The average daily dose (ADD) of exposure for health risk evaluation was estimated by considering few exposure factors as in **Table S7**. For this study, the related age groups were adolescent and adult.

ADD was calculated based on the following ⁴¹ in Eq. 2:

$$ADD (ngkg^{-1}day^{-1}) = \frac{C \times IR \times ED \times EF}{BW \times AT} \quad (2)$$

Where C is the concentration of the heavy metals in air ($ng\ m^{-3}$), IR is the inhalation rate ($m^3\ day^{-1}$), ED is the exposure duration (years), EF is the exposure frequency (days), BW is the body weight (kg) and AT is the averaging time (ED x 365 days).

Non-carcinogenic effects were evaluated by the hazard quotients (HQ) as in Eq. 3:

$$HQ = \frac{ADD}{RfD} \quad (3)$$

Where ADD is the average daily dose and RfD is the reference dose. If the HQ of the chemical is ≤ 1 , there is unlikely risk of developing non-cancer health effects. However, the possibility of non-cancer health effects might occur if the HQ value >1 and there is major possibility of adverse health effects to occur if the HQ value is larger.

2.5.3.2 Health risk assessment for carcinogenic metals

Carcinogenic health risk is defined as the possibility of a person developing any form of cancer from lifetime exposure to carcinogenic threats. The threshold risk is stated as 1×10^{-6} - 1×10^{-4} ³⁸. To evaluate the risk of exposure to carcinogenic metals, excess lifetime cancer risk (ELCR) was considered. The variables used to calculate ELCR were the inhalation unit risk (IUR), slope factor (SF) and the lifetime average daily dose (LADD). LADD was calculated based on the Eq. 2 where AT is the averaging time for cancer risk (70 x 365 days). The details of the parameterisation were reported in Table S7 and S9.

Then, ELCR can be calculated in Eq. 4 and 5 as suggested by EPA³⁸, Peña-Fernández, et al.⁴², Granero and Domingo⁴³:

$$ELCR (inhalation) = LADD \times SF \quad (4)$$

$$SF = IUR \times \left[\frac{1}{IR} \right] \times BW \quad (5)$$

Where LADD is the exposure concentration for inhalation, IR is inhalation rate ($\text{m}^3 \text{ day}^{-1}$) IUR is the unit risk value ($\text{m}^3 \mu\text{g}^{-1}$). Necessary information regarding on carcinogenic types and the unit risks of the metals was obtained from the US EPA Integrated Risk Information System (IRIS).

There were some factors which needed to be evaluated to assess the associated health risk. The factors were chemical elemental composition, the exposed population, the route of exposure and the age-specific groups. The age-specific groups were categorised as infant (0-<1 year),

toddler (1-<6 years), children (6-<12 years), adolescent (12-<18 years) and adult (18-<70 years). As for the route of exposure, possible pathways were via inhalation, ingestion and dermal. Samples for this research were air particle samples, thus would be focusing on inhalation exposure which according to EPA approach, the inhalation exposure estimate was derived in terms of a chronic daily air-intake in (mg/kg per day). The intake of chemical was estimated as a function of few exposure factors, namely the concentration of chemical in the air (CA), inhalation rate (IR), the body weight (BW) and the exposure scenario as mentioned earlier^{37,44}. In this study, the age specific group in interest were adolescents and adults. Since the sampling locations were all educational institutions whereby the students were either adolescents aged within 12-<18 years and also adults aged more than 18 years old. Furthermore, the study area is an overall complex urban with industrial areas nearby. Therefore, there must be vast number of adults who work in the factories and somehow may affect by the unhealthy air condition. Referring to the metals data in PM_{2.5}, a long-term data is required to evaluate the health risk of metals in PM_{2.5}. However, the results obtained in this study will represent for a particular season and the concern of health risk due to the change in the emission of PM_{2.5}-bound metals in the Southern Malaysia region.

2.6 Local weather pattern and transport of air mass

Figure S2 presents the meteorological data plotted in time series. The meteorological data are temperature, relative humidity and wind speed measured at Senai International Airport Station located in Johor, at 1.65 °N, 103.62 °E. These meteorological data was retrieved from Wunderground website⁴⁵. The synoptic wind vector was demonstrated in **Figure 2** retrieving the assimilated data with a resolution of 0.25° × 0.25° from the European Centre for Medium-Range Weather Forecasts (ECMWF) website and plotted them by month from January to

March 2019 using the Grid Analysis and Display System (GrADS). The daily backward trajectories were calculated and cluster by month using Hybrid Single-Particle Lagrangian Integrated Trajectory version 4.9 (HYSPLIT 4.9) windows based compiler⁴⁶. As for the estimation of the backward trajectories using HYSPLIT, the period of backward trajectories is 120 hours. Initially, we calculated 4 trajectories per day on 00.00, 06.00, 12:00 and 18.00 UTC from January 1 to March 31. The estimated all trajectories were used to calculate the cluster of trajectories. All the trajectory data including the two cluster of trajectories were re-plotted using Igor Pro as shown in Figure 2 along with the synoptic wind speed. The model data for HYSPLIT received from <ftp://arlftp.arlhq.noaa.gov/pub/archives/reanalysis>.

3.0 Results and Discussions

3.1 Concentration of trace metals in PM_{2.5}

Table 1 summarizes that for all the monitoring sites, overall the mean concentration of particulate-bound Zn ($29.92 \pm 33.12 \text{ ng/m}^3$) was the highest followed by Se ($27.02 \pm 13.68 \text{ ng/m}^3$) and V ($19.95 \pm 6.99 \text{ ng/m}^3$). Cobalt ($0.06 \pm 0.06 \text{ ng/m}^3$) was least abundant metal among all. Overall, the time series of the metals in PM_{2.5} demonstrated in Figure 3a. At UTHM monitoring site, the three metals with highest mean concentration were Zn ($33.33 \pm 31.22 \text{ ng/m}^3$), Se ($27.64 \pm 15.55 \text{ ng/m}^3$), followed by Cu ($27.39 \pm 21.36 \text{ ng/m}^3$). Concentrations of Zn, Ni and Pb were high in the early period of the monitoring (Figure 3b), with gradual declining trend until the end of monitoring period. Cu reached maximum in February before dropping in March. Both Se and V recorded low concentration in January before reaching peak in March. The trend for mean concentration of trace metals in UTHM was $\text{Co} < \text{Ni} < \text{As} < \text{Pb} < \text{V} < \text{Cu} < \text{Se} < \text{Zn}$. As for SMKGP monitoring site, the mean concentration of Se ($23.07 \pm 14.05 \text{ ng/m}^3$) was found to be the highest, followed by V ($19.58 \pm 8.37 \text{ ng/m}^3$). Concentration of V was relatively higher in February while Ni and Se were notably high in February and

March of the monitoring period (Figure 3c). The trend for mean concentration of trace metals at SMKGP was $\text{Co} < \text{As} < \text{Ni} < \text{Pb} < \text{Cu} < \text{Zn} < \text{V} < \text{Se}$. For PUSPATRI monitoring site, three metals with highest mean concentration were Zn ($48.72 \pm 41.25 \text{ ng/m}^3$), Se ($30.36 \pm 12.83 \text{ ng/m}^3$) and V ($22.01 \pm 6.90 \text{ ng/m}^3$). Concentration of Zn and Pb were high in the beginning of the monitoring period (Figure 3d), with a decline in February before increasing until the end of the monitoring period. The trend was reverse for V where noted with low concentration in January, with a rise in February but gradually declining until the end of the monitoring period. Thus, the trend at PUSPATRI for mean concentration of trace metals was $\text{Co} < \text{Ni} < \text{As} < \text{Cu} < \text{Pb} < \text{V} < \text{Se} < \text{Zn}$. Therefore, Zn was reported as the highest concentration at UTHM and PUSPATRI while Se was the largest in concentration at SMKGP site. Co was the lowest in concentration among all metals in all sites. The change to the concentration of the trace metals is related to the mass concentration of $\text{PM}_{2.5}$ during the study time. A number of factors potentially influence the concentration of $\text{PM}_{2.5}$. Along with the emission sources, the local weather conditions play a great role surging the level of $\text{PM}_{2.5}$. As correlation of trace metals, $\text{PM}_{2.5}$ concentration, and weather variables shown in Table 2, wind speed is negatively correlated with $\text{PM}_{2.5}$ while positive correlation was observed with ambient temperature.

Table 3 summarizes the comparison of trace metals concentration from current monitoring sites to those in other cities from the different parts of the World^{25, 31, 47-54}. Several of the metals (V and Se) in $\text{PM}_{2.5}$ in this study were higher than that observed in other cities including Bangi around Kuala Lumpur in Malaysia. The concentration of $\text{PM}_{2.5}$ -bound V, Cu, Zn, As and Se at present study sites were higher than in neighboring Bangkok and Singapore. Cu, Zn, V and Ni were the most abundant metals for nearly all cities. The coal processing is one of the potential sources of the metals in $\text{PM}_{2.5}$ as reported by Qin, et al.⁵⁵. V and Ni mainly release from the petrochemical oil refinery in Malaysia^{25, 26}. Cu potentially emits from the brake wears as the brake pad consists of Cu reported by Pant⁵⁶. It is presumed that Cu commonly release from brake wears in the urban areas of every cities including the current study sites.

3.2 Influence of local scale weather factors and circulations

From **Figure S2**, it can be observed that the temperature, relative humidity and wind speed kept changing throughout the three-consecutive months (January-March) 2019. In the first week of January, the temperature kept raising from 27.5 °C until 28.8 °C but in the second week, the temperature started declining and then constant in the range of just one-degree change (27.7 ± 1.0 °C). For the third week onwards, the temperature dropped and fluctuated with the lowest temperature was 25.5 °C on 18th of January. In February, the temperature fluctuated but most of the time it was in the range of 27.0 °C to 28.0 °C. The lowest temperature in February was 26.3 °C on 19th of February and the highest was on 14th, which was 28.8 °C. The temperature was mostly high in March. However, in the second week, the temperature dropped slightly and the lowest temperature in March was 26.0 °C and then it rose again. In the time series, the highest temperature recorded was 29.0 °C which were on 13th and 27th of March. The relative humidity in Johor during January to March 2019 was particularly high. Overall, most of the time the relative humidity was in the range of 80-90%. In comparison, the relative humidity was slightly higher in January than in February and March. The highest relative humidity recorded in the time series was on 18th of January, with 94% whereas the least relative humidity was 71%, on 18th of March. In March, relative humidity in Johor was quite decreasing. Next, as for the wind speed, Johor was notably windy in early January and mid-February. The wind speed was quite stable in March. The fastest wind recorded was almost 8mph on 3rd of January and 14th of February. Additionally, the synoptic wind vector was demonstrated in **Figure 2**. It showed that the wind speed was much stronger during January as compared to February and March. While the wind blew from South China Sea, it carried a significant amount of water vapor as well as the pollutants from the mainland of China, leading to change in chemical composition and the concentration largely. As for the correlation among the pollutants and the weather variables as shown in Table 2, wind speed is negatively correlated with PM_{2.5} while positive correlation was observed with ambient temperature. The higher wind

speed enhances the dispersion of air particles such $PM_{2.5}$ causing the lower of $PM_{2.5}$ in the ambient air. The correlation value of ambient temperature and $PM_{2.5}$ is suggested to increase of the concentration of $PM_{2.5}$ in ambient air. However, this is effect is not vibrant to all composition of the air particles, but particular secondary aerosol generates under warming condition. The concentration of trace composition is dependent on the change to the mass concentration of $PM_{2.5}$ in ambient air. A similar observation was reported by Dahari, et al.⁵⁷ in the study area. The correlation values shown in Table 2, the RH is positively related to the mass concentration metals and $PM_{2.5}$. During the wet session in Malaysia (December to March), the RH is relatively higher as compared to the dry session (June to September). In particular, the ionic composition in $PM_{2.5}$ increases during period. However, it is not clear yet in the literature the change of trace metals in $PM_{2.5}$ with respect to the change with RH. An observation was reported in the literature by Sabuti and Mohamed⁵⁸ that the concentration of particulate matters were influenced by monsoon events. During January to March, Northeast monsoon prevailed which affected from the east coast of Malaysian Peninsula as well as the southern part of the South China Sea. During this season, metals such as Ca, Fe, Mn, Cd and Mg were reported with higher concentration than the ambient value.

The backward trajectories were calculated using HYSPLIT 4.9. The cluster of trajectories was re-plotted using Igor Pro as shown in Figure 2. Cluster of the trajectories showed that the air mass was dominantly transporting from the South China Sea. The mixing depth also reportedly demonstrated in a range of 1300 m. Thus, the plume of the pollutants evidently influences in the Malaysian Southern region.

3.3 Sources of metals in $PM_{2.5}$

APCS-MLR was used to obtain quantitatively sources contributing to $PM_{2.5}$ -bound metals. **Table 4** presents the rotated factor loadings via varimax for $PM_{2.5}$ samples. For this study, four factors with significant eigen values were extracted which are vehicular emissions, oil refinery, industrial emissions and undefined. In figure 4(b) the overall site shows percentage of

identified sources are from vehicular emissions (41%), oil refinery and industrial emissions (4%), coal-fired power plant (34%) and undefined mass contribution was 22% to represent unknown sources for metals in PM_{2.5}. The UTHM site from figure 4(c) shows that contribution of identified sources come from vehicle emissions (53%), coal-fired power plant (27%), oil refinery (1%) and undefined sources (19%). Site SMKGP in Figure 4(d) presents contribution of identified sources come from vehicle emissions (14%), coal-fired power plant (45%), oil refinery (7%) and undefined sources (34%). Whereas site PUSPATRI in figure 4(e) presents contribution of identified sources come from vehicle emissions (43%), coal-fired power plant (35%), oil refinery (5%) and undefined sources (18%). Among the three possible sources, vehicle emission was the most predominant for metals for PM_{2.5}. Among the sources of metals in PM_{2.5}, the vehicle emission was identified as the most potential source in UTHM site followed by PUSPATRI and SMKGP. Comparing the estimated total metals (TM) by APCS-MLR and TM determined by ICPMS, the regression line shows a strong correlation ($r^2 = 0.98$) as shown in Figure 4a. Another study conducted in Johor areas also identified vehicular emissions as a key source of PM_{2.5}⁵⁹. Several other studies were also identified the transportation as a potential sources of PM_{2.5} in the Kuala Lumpur and in surrounding areas²⁵⁻²⁷. APCS-MLR results showed that the predicted mass concentration of PM_{2.5}-bound metals correlated well with the PM_{2.5}-bound metals obtained from ICPMS analysis. Thus, the uncertainty of the source apportionment was reasonably lower in this study (Figure 4b).

Factor 1: Vehicle emission

For Factor 1, the high loadings metals were Cu, Zn and Pb. Pb, Cu and Zn were mainly emitted from vehicle emission, as also widely reported in the literature^{60, 61}. Furthermore, Zn was also found accumulated in road dust, mainly released from tires, motor oil and also from the use of motor vehicle brakes. Cu has specific sources like diesel combustion and brake lining wear, as well as from smelting furnace burning⁶² and also emitted as non-exhaust road dust²⁶.

Therefore, Cu, Zn and Pb together indicate Factor 1 and represent emissions from vehicular sources. In Johor, among all other identified sources vehicle emissions emerged as the most potential source of PM_{2.5}-bound metals (Figure 4) contributing 41% of metal concentration. UTHM site shows the largest contribution by vehicle emission.

Factor 2: Oil refinery and industrial emission

The high loadings of Factor 2 were by Co, Ni and As. Ni had been reported coming from oil combustion^{1, 25}. Zhong, et al.⁶³ found that Ni were major emission of various human activities like from industrial processes. Lurie, et al.⁶⁴ reported that Ni were emitted from oil combustion. Shipping emission also release a large amount of Ni as suggested by Cesari, et al.³¹, Cesari, et al.⁶⁵. Thus, Ni were considered as a signature of emissions from oil refinery activities. Miller, et al.⁶⁶ classified Co and As originated from metals industry sources. As emitted from metallurgical processes as supported by Acciai, et al.⁶⁷. Morera-Gómez, et al.⁶⁸ reported that the source of As is waste incineration and Co is common in crustal origin. Since there are quite a few metal industries in Johor, Co and As most probably emit from industrial activities sources. Oil refinery coupled with industrial emission contributed 4% to PM_{2.5}-bound metals. SMKGP site received the largest contribution from this source.

Factor 3: Coal-fired power plant

Se and V was dominantly present in Factor 3. Se is a tracer for coal burning source as reported in the literature^{25, 69, 70}. It was found that there is a coal-fired plant in Johor Bahru in a radius within 30 km from SMKGP sampling station. Hence, traces of Se might originated from coal-fired plant. This source has potentially contributed 34% to the PM_{2.5}-bound metals.

3.3 Human exposure of the toxic metals in PM_{2.5}

3.3.1 Non-carcinogenic metals effect

In **Table S8**, in overall ADD Zn has the highest mean value for both adolescent and adult. The sequence of the metals by their mean ADD values for overall site are $\text{Co} < \text{Ni} < \text{As} < \text{Pb} < \text{Cu} < \text{V} < \text{Se} < \text{Zn}$ for both age groups. For site UTHM, the mean ADD value sequence are $\text{Co} < \text{Ni} < \text{As} < \text{Pb} < \text{V} < \text{Se} < \text{Cu} < \text{Zn}$ for both age groups. For site SMKGP, the mean ADD value sequence are $\text{Co} < \text{As} < \text{Ni} < \text{Pb} < \text{Cu} < \text{Zn} < \text{V} < \text{Se}$ for both age groups. Whereas for site PUSPATRI, the mean ADD value sequence are $\text{Co} < \text{Ni} < \text{As} < \text{Cu} < \text{Pb} < \text{V} < \text{Se} < \text{Zn}$ for both age groups. From the ADD values above, the hazard quotient (HQ) was calculated using the RfD values from **Table S9**. From **Table 5**, all metals have HQ values < 1 , thus there is small possibilities for non-cancer health risks to occur. The highest hazard quotient for both adolescent and adult was reported from As and the lowest was from Co. Hence, As was comparatively potentially more hazardous to human health as compared to other metals. At overall site, UTHM, SMKGP and PUSPATRI site, the sequence of the metals by their mean HQ values are $\text{Co} < \text{Zn} < \text{Cu} < \text{Pb} < \text{Ni} < \text{As}$ for both age group. MohseniBandpi, et al.⁷¹ reported that the HQ trend values were $\text{Mn} > \text{Cr} > \text{As} > \text{Pb} > \text{Cd} > \text{V} > \text{Cu} > \text{Ni} > \text{Zn}$ which the study was done in urban areas of Tehran, Iran during summertime. This showed that the result matched current study whereby As with the highest HQ value and Zn among the lowest HQ value. From the above graphical figure, for all metals, HQ for adolescent was higher than for adult. The highest HQ value for both adult and adolescent was As, with adolescent's value two times higher than the adult, whereby the adult's average HQ value for As was 1.63×10^{-2} and for adolescent it was 3.00×10^{-2} . Although the values were still under the risk (< 1), the adolescent whom in this study were students shall be more aware of their health.

3.3.2 Carcinogenic metals effect

From Table S10, Zn was reported with the highest mean LADD value for both adolescent and adult age groups. The sequence of the metals for overall site by their mean LADD values are the same for both age groups which is $\text{Co} < \text{Ni} < \text{As} < \text{Pb} < \text{Cu} < \text{V} < \text{Se} < \text{Zn}$. The sequence of the metals for UTHM site by their mean LADD values are the same for both age groups which is $\text{Co} < \text{Ni} < \text{As} < \text{Pb} < \text{V} < \text{Cu} < \text{Se} < \text{Zn}$. Sequence of the metals for SMKGP site by their mean LADD values are the same for both age groups which is $\text{Co} < \text{As} < \text{Ni} < \text{Pb} < \text{Cu} < \text{Zn} < \text{V} < \text{Se}$. Whereas the sequence of the metals for PUSPATRI site by their mean LADD values are the same for both age groups which is $\text{Co} < \text{Ni} < \text{As} < \text{Cu} < \text{Pb} < \text{V} < \text{Se} < \text{Zn}$. From the LADD values, the excess lifetime cancer risk (ELCR) was calculated using the IUR, SF, IR and BW values listed in **Table S7 and S9**. As shown in Table 6, the ELCR values of both adolescent and adult age groups were within the acceptable range. Among the three metals, ELCR value for As was comparatively higher than Ni and Pb for both adult and adolescent age groups. The metals with the least ELCR value were Pb. The sequence for the mean ELCR values of the overall site, UTHM, SMKGP and PUSPATRI site are in the same order of $\text{Pb} < \text{Ni} < \text{As}$ for both age group. Reported by Sulong, et al. ²⁷, it was observed the sequence are $\text{Cr} > \text{As} > \text{Co} > \text{Ni} > \text{Cd} > \text{Pb}$ trend for ELCR, which correlated with the present study whereby Pb had lower ELCR value than Ni. From the above figure, in comparison, the ELCR value for As was significantly higher than other metals. Adult was more affected than adolescent age group for cancer risk assessment from the ELCR values. Several other studies also reported the adult with higher potential of cancer risk compared to other age groups for instance in Kuala Lumpur, Malaysia ²⁷ and in Delhi City, India ⁴⁰.

4.0 Conclusions

In this study, Zn was detected with the highest average concentration whereas Co with the lowest concentration in $\text{PM}_{2.5}$ samples from Johor among the metals effectively analysed. The

concentration of heavy metals in PM_{2.5} samples were affected significantly by the ambient temperature, wind speed and also relative humidity (RH). This study observed that the higher wind speed enhances the dispersion of air particles such PM_{2.5} causing the lower of PM_{2.5} in the ambient air. The correlation value of ambient temperature and PM_{2.5} suggests an increase of the concentration of PM_{2.5} in ambient air as the temperature increases. However, this is effect is not vibrant to all composition of the air particles, but particular secondary aerosol. The source apportionment by APCS model identified three main sources i.e. vehicle emission, oil refinery activity and coal fired plant. The vehicle emission was the predominant among other identified sources of metals in PM_{2.5}. From the health risk assessment, the metals were reported within the tolerance levels for HQ and ELCR values, respectively for both age groups in interest, which were adolescent and adult. For non-carcinogenic metals effect, HQ values for all metals were higher for adolescent compared to adults. Alternatively, for carcinogenic metals effect, the adult age group was reported with higher ELCR values for all the metals than adolescent. For both adolescent and adult, the sequence of the metals by their mean HQ values was Co < Zn < Cu < Pb < Ni < As while the sequence of the metals by their mean ELCR values was Pb < Ni < As, also for both of the age groups. The results show that the As was the most potentially toxic element for both age groups but the adolescent are more vulnerable to non-carcinogenic risk compared to adults. However, the adults were more affected than adolescent for cancer related health effects. Even though the hazard and risk characterizations for all metals were still within the acceptable limit, the possible effects in long term exposure shall not be neglected, especially for the metals like As and Ni with high values of HQ and ELCR such as Ni and As especially. Thus, to prevent the condition of ambient air from worsening, the people must be aware and take necessary actions. For instance, citizens who work in the same place can practice carpooling which is also economical. Next, it is advised to use the public transportations available more efficiently. A single, irresponsible action will have a huge

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3 impact if it is done by many individuals hence, the least thing every person could do is by avoid
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5 contributing to air pollution such as stop open burning and reduce unnecessary vehicle trips by
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7 limiting outdoor activities.
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10 11 12 13 14 **Acknowledgement**

15
16 This research is supported by University of Malaya Research Grant IIRG009A-2019, and
17
18 Fundamental Research Grant Scheme (FRGS) FP099-2019A. We also sincerely acknowledge
19
20 the Department of Environment (DOE) and Pakar Scieno Trans Water for providing us the
21
22 filter samples. Tirthankar Banerjee acknowledges fund received under ASEAN- India
23
24 Collaborative Research and Development Scheme (CRD/2018/000011). Our appreciations
25
26 also go to the science officers in Chemistry Department in the University of Malaya for
27
28 assisting and guiding the analysis while using the facilities and instrument provided by the
29
30 department. The data of the Hysplit model were available at [ftp://arlftp.arlhq.](ftp://arlftp.arlhq.noaa.gov/pub/archives/reanalysis)
31
32 [noaa.gov/pub/archives/reanalysis](http://arlftp.arlhq.noaa.gov/pub/archives/reanalysis). The authors gratefully acknowledge the NOAA Air
33
34 Resources Laboratory for the provision of the Hysplit transport and dispersion model and/or
35
36 READY website (<http://www.ready.noaa.gov>) used in this publication.
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Tables

Table 1. Summary statistics of the metals in PM_{2.5}

Overall	Unit	N	Mean	GM	Median	Min	Max	10%	90%	SD
V	ng/m ³	18	19.65	18.32	19.40	7.56	34.23	7.95	28.91	6.99
Co	ng/m ³	14	0.06	0.03	0.05	0.00	0.20	0.00	0.20	0.06
Ni	ng/m ³	18	2.46	1.88	1.72	0.52	6.81	0.84	5.80	1.96
Cu	ng/m ³	18	13.39	8.25	6.99	2.22	53.01	2.35	47.93	15.66
Zn	ng/m ³	18	29.92	16.78	12.85	0.87	112.41	3.51	94.30	33.12
As	ng/m ³	18	3.79	2.90	2.97	0.99	14.76	1.03	7.54	3.35
Se	ng/m ³	18	27.02	23.16	29.94	8.41	46.59	9.95	42.97	13.68
Pb	ng/m ³	18	7.75	4.97	4.33	1.30	33.60	1.45	22.48	8.57
Total metals	ng/m ³	18	104.04	92.81	86.82	38.64	209.64	50.71	193.46	51.83
PM _{2.5}	μg/m ³	18	27.98	27.44	27.70	18.20	38.00	20.30	37.70	5.66
T	°C	18	27.37	27.35	27.69	26.00	28.83	26.00	28.61	1.03
RH	%	18	84.11	83.92	83.40	75.90	92.00	75.90	92.00	5.69
WS	mph	18	4.66	4.44	4.80	2.80	6.80	2.80	6.80	1.46
UTHM										
V	ng/m ³	6	17.35	16.30	16.83	7.56	24.20	7.56	24.20	5.96
Co	ng/m ³	3	0.04	0.03	0.05	0.01	0.07	0.01	0.07	0.03
Ni	ng/m ³	6	1.38	1.29	1.11	0.84	2.27	0.84	2.27	0.59

Cu	ng/m ³	6	27.39	17.77	24.62	2.35	53.01	2.35	53.01	21.36
Zn	ng/m ³	6	33.33	25.08	23.23	11.52	94.30	11.52	94.30	31.22
As	ng/m ³	6	3.07	2.83	2.58	1.67	5.05	1.67	5.05	1.39
Se	ng/m ³	6	27.64	23.48	29.52	9.95	42.97	9.95	42.97	15.55
Pb	ng/m ³	6	11.50	7.22	7.00	1.30	33.60	1.30	33.60	11.88
Total metals	ng/m ³	6	121.69	112.89	111.82	62.34	209.64	62.34	209.64	52.27
PM _{2.5}	µg/m ³	6	27.45	26.59	24.75	18.20	37.70	18.20	37.70	7.62
SMKGP										
V	ng/m ³	6	19.58	17.80	20.77	7.95	28.91	7.95	28.91	8.37
Co	ng/m ³	6	0.05	0.02	0.03	0.00	0.20	0.00	0.20	0.07
Ni	ng/m ³	6	3.23	2.66	2.60	1.07	5.80	1.07	5.80	2.11
Cu	ng/m ³	6	5.90	5.13	5.41	2.22	10.93	2.22	10.93	3.26
Zn	ng/m ³	6	7.69	5.69	9.21	0.87	12.00	0.87	12.00	4.63
As	ng/m ³	6	2.29	2.07	2.32	0.99	3.31	0.99	3.31	1.04
Se	ng/m ³	6	23.07	19.20	21.46	8.41	38.66	8.41	38.66	14.05
Pb	ng/m ³	6	4.26	3.87	4.15	1.88	7.39	1.88	7.39	1.95
Total metals	ng/m ³	6	66.08	63.03	62.03	38.64	95.77	38.64	95.77	21.89
PM _{2.5}	µg/m ³	6	26.28	25.98	26.70	20.30	31.00	20.30	31.00	4.32
PUSPATRI										
V	ng/m ³	6	22.01	21.19	20.14	14.01	34.23	14.01	34.23	6.90
Co	ng/m ³	5	0.09	0.06	0.08	0.01	0.20	0.01	0.20	0.07
Ni	ng/m ³	6	2.77	1.95	1.65	0.52	6.81	0.52	6.81	2.46
Cu	ng/m ³	6	6.89	6.15	5.08	3.41	12.79	3.41	12.79	3.73
Zn	ng/m ³	6	48.72	33.11	41.49	9.21	112.41	9.21	112.41	41.25
As	ng/m ³	6	6.00	4.17	5.41	1.03	14.76	1.03	14.76	5.10
Se	ng/m ³	6	30.36	27.55	31.35	11.05	46.59	11.05	46.59	12.83
Pb	ng/m ³	6	7.51	4.40	3.02	1.45	22.48	1.45	22.48	8.55
Total metals	ng/m ³	6	124.33	112.35	123.79	56.44	193.46	56.44	193.46	57.66
PM _{2.5}	µg/m ³	6	30.22	29.92	29.45	24.20	38.00	24.20	38.00	4.74

Table 2. Spearman Rank Order correlation among the variables in PM_{2.5} with meteorological factors

	V	Co	Ni	Cu	Zn	As	Se	Pb	TM	PM _{2.5}	T	RH	WS
V	1.000												
Co	0.039	1.000											
Ni	.476*	0.261	1.000										
Cu	0.018	-0.019	0.003	1.000									
Zn	-0.133	0.053	-0.381	0.465	1.000								
As	0.381	0.376	0.356	0.042	0.247	1.000							
Se	.484*	-0.102	-0.137	-0.271	0.096	0.412	1.000						
Pb	-0.042	0.078	0.139	.833**	.503*	0.104	-0.313	1.000					
TM	0.298	-0.028	-0.063	.482*	.711**	.503*	.513*	.527*	1.000				
PM _{2.5}	.676**	-0.090	0.261	0.243	0.166	0.323	0.331	0.181	0.346	1.000			
T	0.275	0.188	-0.204	0.095	0.101	0.091	-0.031	-0.002	0.041	-0.078	1.000		
RH	-0.378	-0.135	0.069	-0.028	0.011	0.001	0.025	0.086	0.039	0.016	-.911**	1.000	
WS	-0.048	0.179	-0.019	-0.085	-0.155	-0.302	-0.413	-0.142	-0.402	-0.372	.513*	-.658**	1.000

*Correlation is significant at $p < 0.05$; **significant at $p < 0.01$; TM: total metals

Table 3: Comparison of metals concentration (ng/m³) in PM_{2.5} from the current study and other cities in different parts of the world

Cities	V	Co	Ni	Cu	Zn	As	Se	Pb	References
Bangkok, Thailand	2.6		1.6	4.4	34	1.2		9.7	Jariya Kayee et al. 2020
*Singapore					0.718	0.017	0.01	0.283	George et al. 2020
Jakarta, Indonesia	3.92	3.52	2.47	5.67	75.06			40.2	Santoso et al. 2013
Bangi, Malaysia	5.13	0.85	17.24	28.33	389.2	5.76	0.65	21.84	Khan et al. 2016
Ho Chi Minn, Vietnam			32	391	128			225	Phan et al. 2020
Shenzhen, China	11.59	2.2	4.76	17.32		6.98		31.02	Qin et al., 2020
Beijing, China	5.8	1.2		136.7	292.6			206.3	Lin et al. 2020
Brindisi, Italy	3		2.5	2.4	16.3			4.4	Cesari et al. 2014
Birmingham, UK	1.2			95.6	93.4				Pant et al. 2017
Brisbane, Australia	0.8	0.7	0.5	2	15.5			5	Friend et al., 2011
Kolkata, India	9.5	2.1	40	58	542			368	Das et al., 2015
Johor, Malaysia	19.65	0.06	2.46	13.39	29.92	3.79	27.02	7.75	This study

Unit: ng/m³, *PM_{2.0-4.0}

Table 4: PCA factor loadings and eigenvalues of metals in PM_{2.5} samples

Variables	Factor 1	Factor 2	Factor 3
V	-0.204	0.404	0.595
Co	-0.003	0.704	-0.306
Ni	-0.048	0.832	0.129
Cu	0.704	-0.357	-0.313
Zn	0.903	0.113	0.255
As	0.387	0.687	0.398
Se	-0.045	-0.145	0.926
Pb	0.839	0.148	-0.318
Eigen value	2.658	2.268	1.481
Variance (%)	31.434	26.826	17.520
Cumulative (%)	31.434	58.260	75.780
Identified sources	Vehicle emission	Oil refinery and industries	Coal fired plants

Table 5. Summary table of hazard quotient (HQ) for adolescent and adults

HQ	Metals	Valid N	Mean	Minimum	Maximum	Std.Dev.	
Overall (Johor)	HQ (adolescent)	Co	15	1.169E-07	0.000E+00	3.950E-07	1.271E-07
		Ni	18	4.875E-03	1.025E-03	1.350E-02	3.887E-03
		Cu	18	1.326E-04	2.202E-05	5.251E-04	1.551E-04
		Zn	18	3.951E-05	1.149E-06	1.485E-04	4.373E-05
		As	18	3.001E-02	7.869E-03	1.169E-01	2.656E-02
		Pb	18	8.778E-04	1.468E-04	3.803E-03	9.704E-04
	HQ (Adults)	Co	18	5.29E-08	0.00E+00	2.14E-07	6.72E-08
		Ni	18	2.65E-03	5.57E-04	7.33E-03	2.11E-03
		Cu	18	7.20E-05	1.20E-05	2.85E-04	8.42E-05
		Zn	18	2.14E-05	6.24E-07	8.06E-05	2.37E-05
		As	18	1.63E-02	4.27E-03	6.35E-02	1.44E-02
		Pb	18	4.77E-04	7.97E-05	2.06E-03	5.27E-04
UTHM	HQ (adolescent)	Co	3	8.36E-08	1.05E-08	1.39E-07	6.62E-08
		Ni	6	2.73E-03	1.66E-03	4.49E-03	1.18E-03
		Cu	6	2.71E-04	2.33E-05	5.25E-04	2.12E-04
		Zn	6	4.40E-05	1.52E-05	1.25E-04	4.12E-05
		As	6	2.44E-02	1.33E-02	4.00E-02	1.10E-02
		Pb	6	1.30E-03	1.47E-04	3.80E-03	1.35E-03
	HQ (Adults)	Co	6	2.27E-08	0.00E+00	7.56E-08	3.37E-08
		Ni	6	1.48E-03	9.01E-04	2.44E-03	6.38E-04
		Cu	6	1.47E-04	1.26E-05	2.85E-04	1.15E-04
		Zn	6	2.39E-05	8.26E-06	6.76E-05	2.24E-05
		As	6	1.32E-02	7.20E-03	2.17E-02	5.98E-03
		Pb	6	7.07E-04	7.97E-05	2.06E-03	7.30E-04
SMKGP	HQ (adolescent)	Co	6	1.09E-07	8.27E-09	3.90E-07	1.47E-07
		Ni	6	6.40E-03	2.11E-03	1.15E-02	4.17E-03
		Cu	6	5.84E-05	2.20E-05	1.08E-04	3.23E-05
		Zn	6	1.02E-05	1.15E-06	1.58E-05	6.12E-06
		As	6	1.81E-02	7.87E-03	2.62E-02	8.25E-03
		Pb	6	4.82E-04	2.13E-04	8.36E-04	2.21E-04
	HQ (Adults)	Co	6	5.91E-08	4.49E-09	2.12E-07	7.96E-08
		Ni	6	3.48E-03	1.15E-03	6.24E-03	2.26E-03
		Cu	6	3.17E-05	1.20E-05	5.88E-05	1.76E-05
		Zn	6	5.52E-06	6.24E-07	8.60E-06	3.32E-06
		As	6	9.85E-03	4.27E-03	1.42E-02	4.48E-03
		Pb	6	2.62E-04	1.15E-04	4.54E-04	1.20E-04
PUSPATRI	HQ (adolescent)	Co	6	1.42E-07	0.00E+00	3.95E-07	1.43E-07
		Ni	6	5.48E-03	1.03E-03	1.35E-02	4.88E-03
		Cu	6	6.82E-05	3.38E-05	1.27E-04	3.70E-05
		Zn	6	6.43E-05	1.22E-05	1.48E-04	5.45E-05
		As	6	4.75E-02	8.17E-03	1.17E-01	4.04E-02
		Pb	6	8.50E-04	1.64E-04	2.54E-03	9.68E-04
	HQ (Adults)	Co	6	7.69E-08	0.00E+00	2.14E-07	7.76E-08
		Ni	6	2.98E-03	5.57E-04	7.33E-03	2.65E-03
		Cu	6	3.70E-05	1.83E-05	6.88E-05	2.01E-05
		Zn	6	3.49E-05	6.60E-06	8.06E-05	2.96E-05
		As	6	2.58E-02	4.44E-03	6.35E-02	2.19E-02
		Pb	6	4.61E-04	8.92E-05	1.38E-03	5.26E-04

Table 6. Summary table of excess lifetime cancer risk (ELCR) for adolescent and

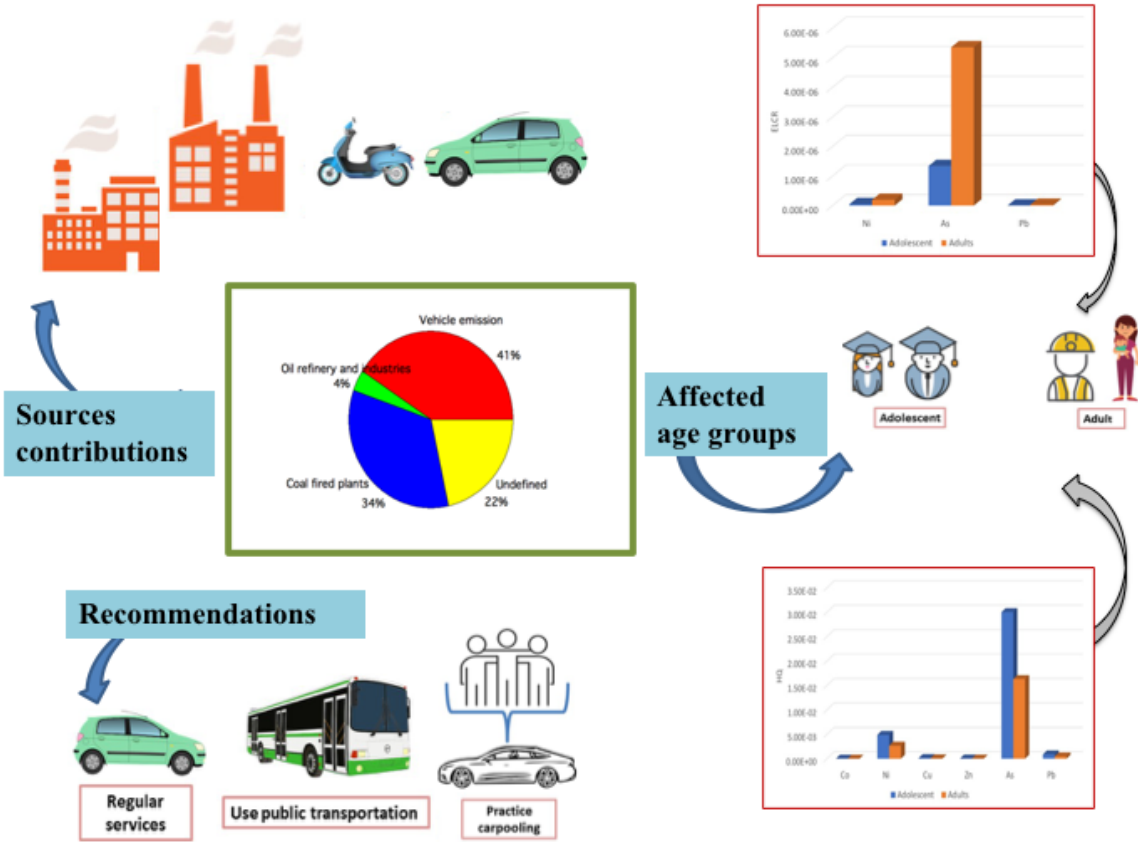
ELCR		Valid N	Mean	Minimum	Maximum	Std.Dev.
Overall (Johor)	ELCR (adol.)	Ni	18	4.85E-08	1.02E-08	1.34E-07
		As	18	1.34E-06	3.51E-07	5.22E-06
		Pb	18	7.65E-09	1.28E-09	3.31E-08
	ELCR (adul.)	Ni	18	1.94E-07	4.08E-08	5.38E-07
		As	18	5.35E-06	1.40E-06	2.09E-05
		Pb	18	3.06E-08	5.12E-09	1.33E-07
UTHM	ELCR (adol.)	Ni	6	2.72E-08	1.65E-08	4.47E-08
		As	6	1.09E-06	5.92E-07	1.79E-06
		Pb	6	1.13E-08	1.28E-09	3.31E-08
	ELCR (adul.)	Ni	6	1.09E-07	6.61E-08	1.79E-07
		As	6	4.35E-06	2.37E-06	7.14E-06
		Pb	6	4.54E-08	5.12E-09	1.33E-07
SMKGP	ELCR (adol.)	Ni	6	6.38E-08	2.10E-08	1.15E-07
		As	6	8.09E-07	3.51E-07	1.17E-06
		Pb	6	4.20E-09	1.85E-09	7.29E-09
	ELCR (adul.)	Ni	6	2.55E-07	8.41E-08	4.58E-07
		As	6	3.24E-06	1.40E-06	4.68E-06
		Pb	6	1.68E-08	7.41E-09	2.91E-08
PUSPATRI	ELCR (adol.)	Ni	6	5.46E-08	1.02E-08	1.34E-07
		As	6	2.12E-06	3.65E-07	5.22E-06
		Pb	6	7.40E-09	1.43E-09	2.22E-08
	ELCR (adul.)	Ni	6	2.18E-07	4.08E-08	5.38E-07
		As	6	8.48E-06	1.46E-06	2.09E-05
		Pb	6	2.96E-08	5.73E-09	8.87E-08

Adol.: adolescent; adul.: adults

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37 Graphical abstract



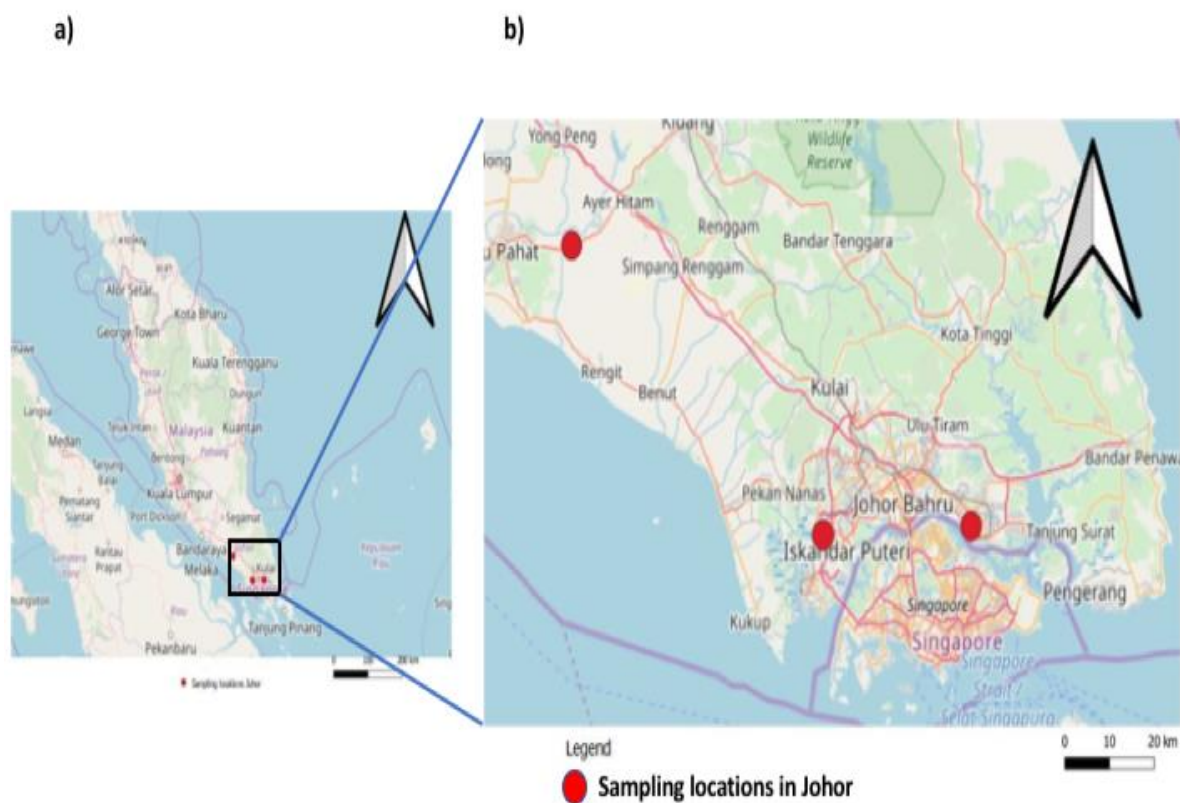
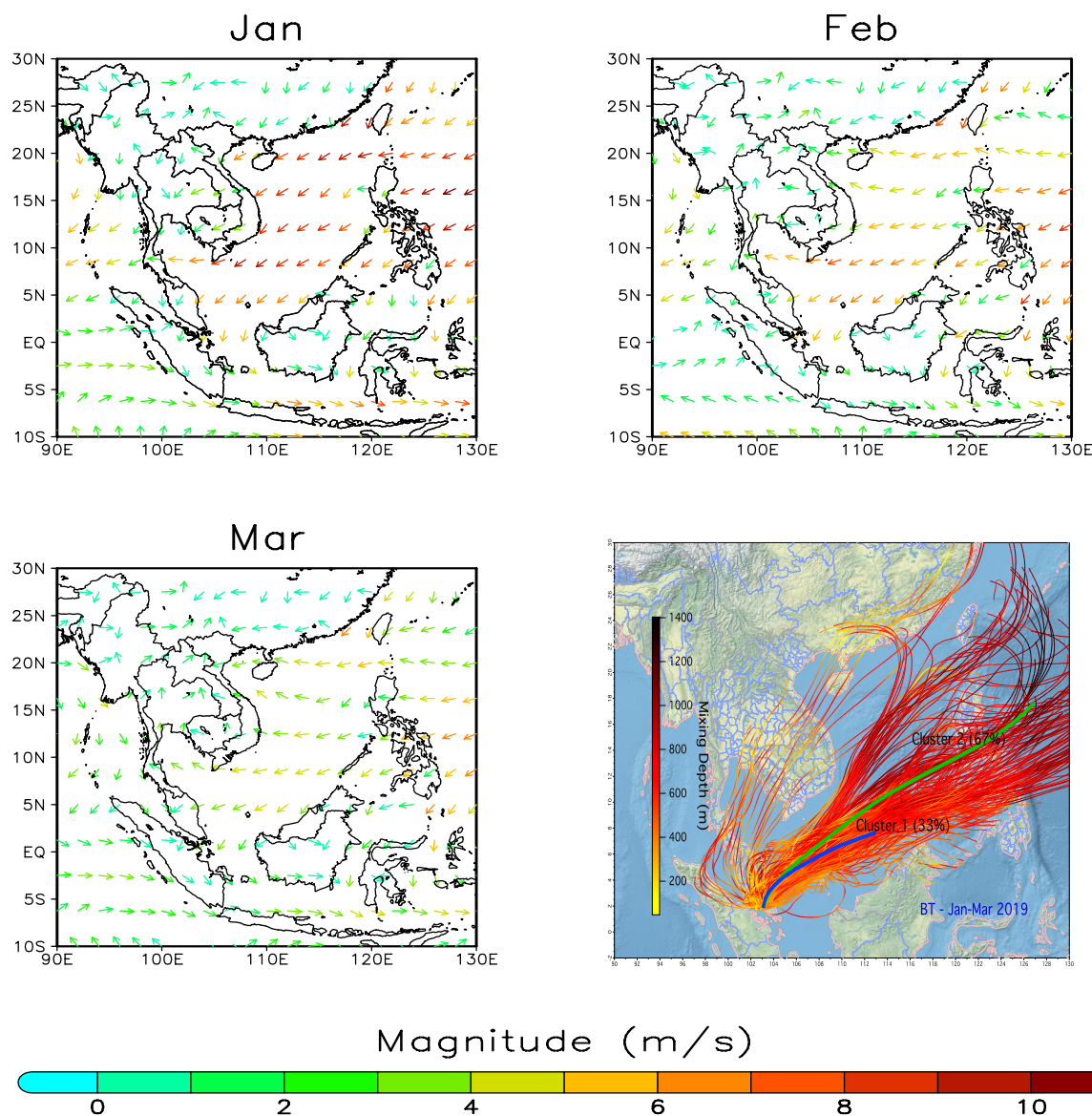


Figure 1: Study locations a) Johor state in Malaysian Peninsula and b) sampling sites in Johor



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52 **Figure 2:** A synoptic scale wind vector and cluster of backward trajectories over
53 Southeast Asia during January to March, 2019.

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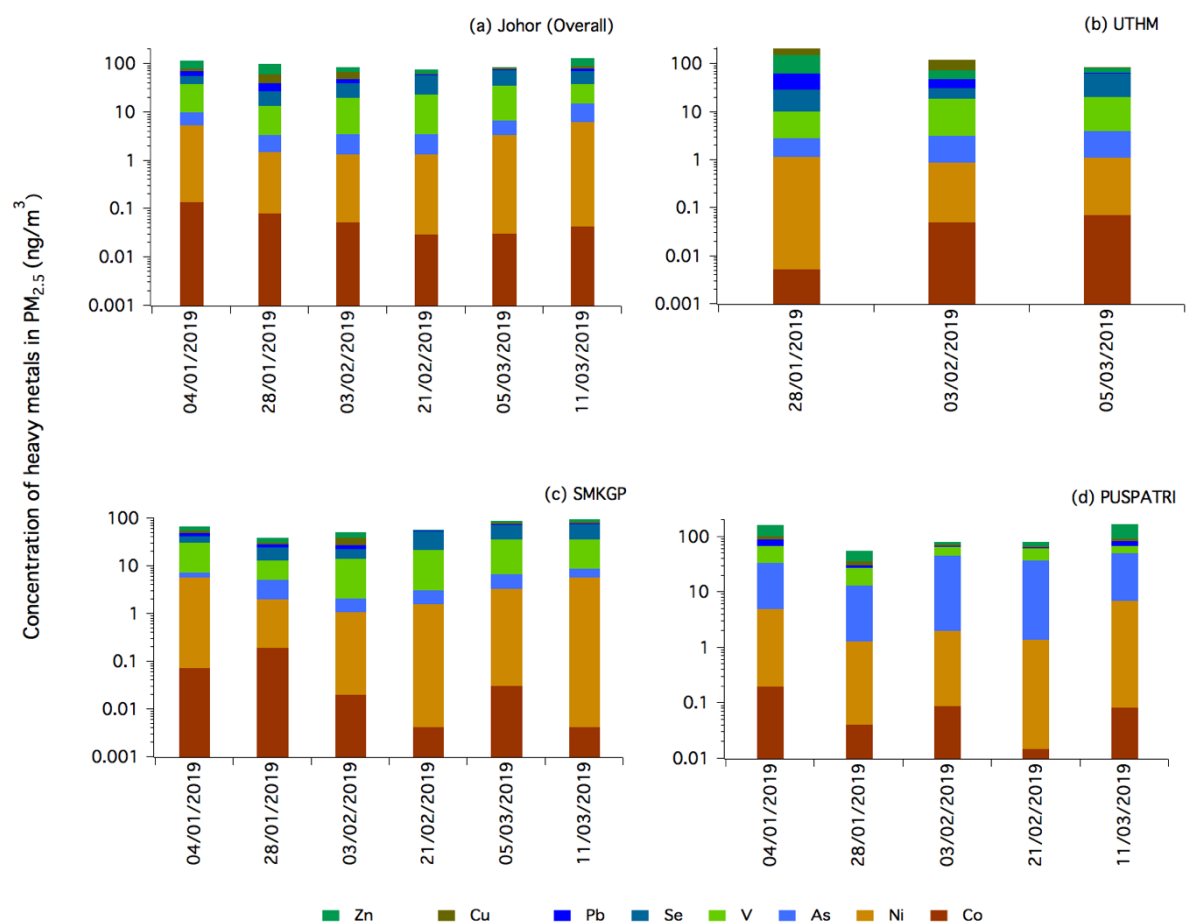


Figure 3 Time series of the heavy metals (a) overall Johor, (b) UTHM, (c) SMKGP and (d) PUSPATRI.

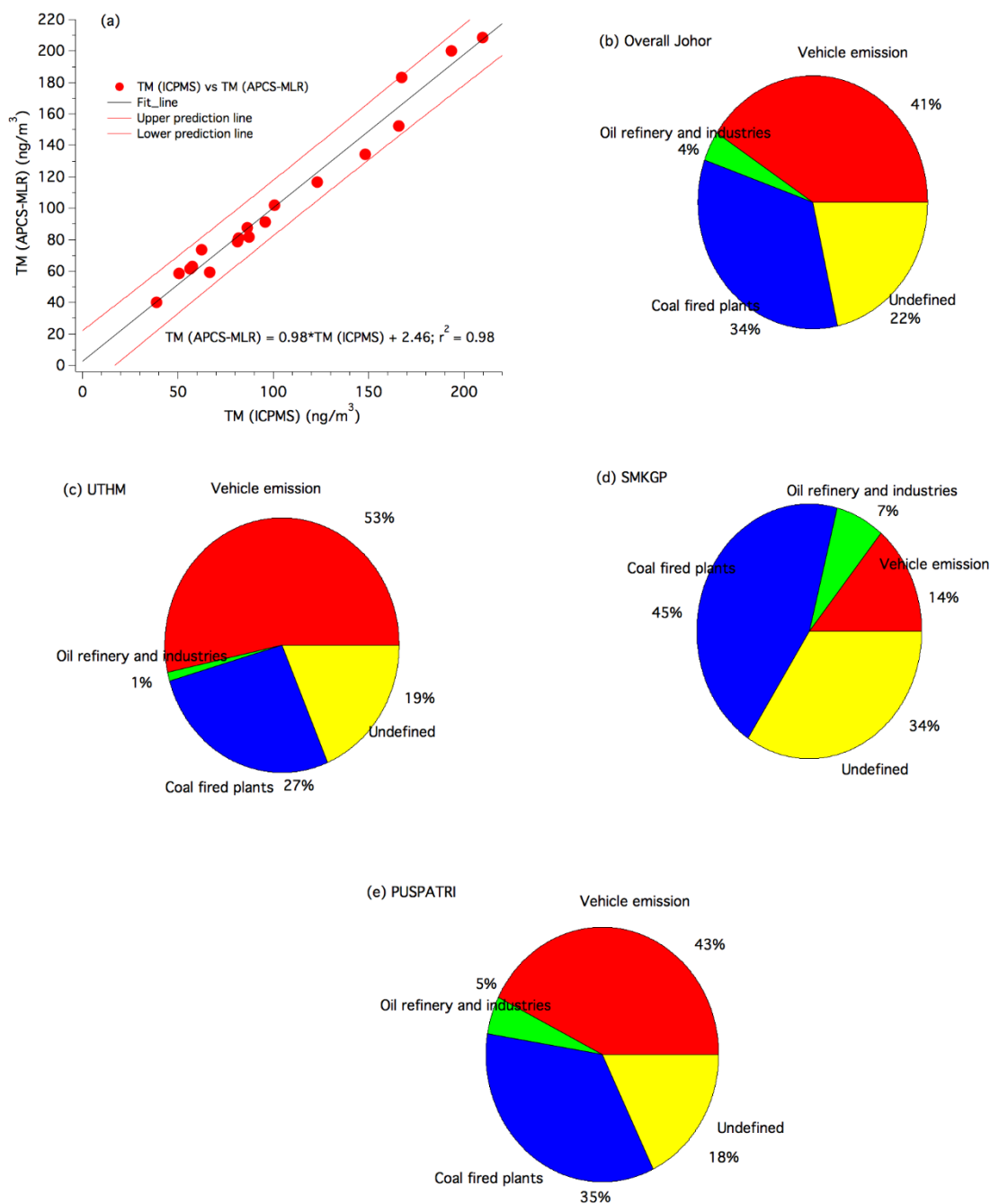


Figure 4: Contribution of the identified sources for metals in PM_{2.5} by APCS-MLR.