Concentration of the PM$_{2.5}$ according to the sampling areas associated with the concentration of heavy metals and anions in Penang

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Abstract

Particulate matter (PM) was identified as the main air pollutants in Malaysia which directly impose a threat to human health and understanding this pollutant is also crucial for its mitigation and control. This study aims to determine and characterize the PM$_{2.5}$ with the influence of meteorological parameters. The PM$_{2.5}$ samples were collected at Perai, Nibong Tebal and highway of Penang Bridge represented the industrial, residential, and roadside areas of Mainland Penang, respectively. The descriptive statistical analysis was conducted to understand the variations of PM$_{2.5}$ concentration on the study areas, while its relationship with meteorological parameters were tested using Pearson correlation. Eleven heavy metals and three anions were determined using mass spectrometry (ICP-MS) and Ion Chromatography, respectively. Results showed that the mean concentration of PM$_{2.5}$ are 10.88 μg/m³, 31.96 μg/m³, and 39.35 μg/m³ for residential area, industrial area, and roadside, respectively. The results revealed that mean concentrations of PM$_{2.5}$ (39.35 μg/m³) in the roadside are significantly higher than NAAQG 24-h average of 35 μg/m³. Result also suggested that meteorological parameters were least significant in controlling the PM$_{2.5}$ concentrations as only the air pressure was found to be significant with the value of correlation coefficient of 0.476. The major components such as SO$_4^{2-}$, K and Na account for about 70-90% of the PM$_{2.5}$.

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1. INTRODUCTION

Air pollution has been a substantial challenge among the various environmental problems to affect the biggest damage to health and loss of welfare from environmental causes in Asian countries (Dominick et al., 2012). Particulate matter (PM) is one of the atmospheric pollutants that has been an environmental and health concern in many developed countries (Sun et al., 2015). In Malaysia, PM also identified as the main air pollutants of concern and two type PM which as particulate matter with aerodynamic diameter less than 10 micron (PM$_{10}$) and particulate matter with aerodynamic diameter less than 2.5 micron (PM$_{2.5}$) were this country criteria air pollutants. However, the fine portion of atmospheric particulate matter received higher attention as it imposes higher risk towards human health. As it very small in size, Figueroa et al. (2006) reported that PM$_{2.5}$ have higher capacity to enter human lung and deep deposited in the lung. Some of the factors that influence water quality and it’s chemistry across African coast are enhanced by Na$^+$, Ca$^{2+}$, Mg$^{2+}$ and HCO$_3^-$, due to silicate and carbonate weathering and enhanced NO$_3^-$ from indiscriminate waste disposal from human activities (Wotany et al., 2013). The chemical quality of surface and groundwater is controlled by natural processes such as precipitation, mineral weathering and evaporation-crystallization and significantly influenced by anthropogenic processes (Wotany et al., 2013; Mangoua et al., 2015).

Commonly, similarly to PM, the PM$_{2.5}$ is a compound of horizons as it contains variety of solid and liquid particles with size smaller than 2.5 micrometer. PM$_{2.5}$ also released to the atmosphere by natural and man-made sources such as combustions, vehicular and industrial emissions as well other activities such as mining and constructions. The chemical composition of PM$_{2.5}$ offers valuable information to identify the contributions of specific sources, and to understand aerosol properties and
processes that could affect health, climate, and atmospheric conditions (Solomon et al., 2014; Fatimah, 2016).

USEPA reported that the major components of PM$_{2.5}$ are ammonium sulphate, ammonium nitrate, organic carbonaceous mass, elemental carbon, and crustal material which included heavy metals. Heavy metals in PM$_{2.5}$ that can be breathed into the lung show a rise lung or cardiopulmonary damages (Lee and Hieu, 2011). Heavy metals invade the environment via three main paths such as deposition of particulates, disposal of metals via sewages and by-product from metal mining operation. In addition, biogeochemical cycle is the major phase of heavy metals as they are absorbed to PM$_{2.5}$ in the atmosphere. They also can travel far away from its original position and unload to the ground through wet and dry deposition, resulting a risk to the environment (Li et al., 2015). For the coarse particles, the ions and its heavy metals exist are sulphate (SO$_4^{2-}$), chloride (Cl$^-$), nitrate (NO$_3^-$), iron (Fe), potassium (K) and sodium (Na) whereas for the fine particles, the ions and its heavy metals are SO$_4^{2-}$, K and Na (Tahir et al., 2013).

The continuous developments in Penang have raised an important role of PM$_{2.5}$ and its associates in the atmosphere. High emissions of PM$_{2.5}$ has a strong potential for adsorbing environmental toxicants due to the large specific surface area (Sun et al., 2015). According to Predeep (2015), Penang has reached near to the level of unhealthy zone between 100 to 200 based on the API reading in 2013. This problem is the severe issue to be solved and considered by societies especially the authorities. The objective of this study is to determine PM$_{2.5}$ concentrations and their associated selected heavy metals and anion ions in the samples collected from the study areas. It is prerequisite to understand the chemicals composition and concentration of PM$_{2.5}$ of sample area which caused deterioration in human health and environment. The predictable study of PM$_{2.5}$ and its associated chemicals may deliver crucial information of the origin and sources of PM$_{2.5}$ which is vital to invest precautions to decrease pollution in the current sampling area.

2. MATERIALS AND METHODS

2.1. Study Area

PM$_{2.5}$ samples was collected from three sampling areas namely Perai, highway of Penang Bridge, and Nibong Tebal representing industrial area, roadside area and residential area of Penang mainland, respectively from June until September 2015. Perai (5°23.4704N; 100°23.1977E) has high density of industrial and medium density of residential dwellings with the most of air pollutants originated from industrial zone (Zakaria, 2007). It was nominated as station 1 (S1). Second station (S2) was Nibong Tebal (5°10’5″N 100°28’43″E) and can be considered as suburban area with a huge number of populations representing as residential areas (Shahnon, 2005). Highway of Penang Bridge was notified as Station 3 (S3) and the sampler was placed at Toll Office near to Penang Bridge (5° 21.375 N 100° 23.584 E). A total of nine samples consisting of three samples of each location were collected monthly. Due to a limited number of samplers, the samples were collected simultaneously representing all the three different locations. Duration of sampling was 24 hours per day that was taken from first, middle, and the last week of each month from June to September 2015. The total number of samples collected in these four months was 36. The location of the sampling sites in Penang shown in Figure 1.

2.2. Data Acquisition

The materials used throughout this study were quartz filter paper with 25 mm in diameter by using the instrument named Low Volume Air Sampler (LVAS) with flow rate of 20 l/min for period of 24 hours. The quartz microfilter were kept under controlled conditions in the desiccator before and after sampling at relative humidity of 50 ± 5% for about 48 hours in order to minimize the influence of water adsorption. The filter papers were weighed using the 5-digit microelectronic balance with 0.01 mg sensitivity before and after sampling. Daily average meteorological parameters data from June until September 2015 were obtained from the Meteorological Department of Penang. The meteorological parameters included surface temperature, relative humidity and precipitation, wind speed, wind direction, and air pressure.

2.3. Data Analysis

The collected data then were analysed using Pearson correlation with the aid of SPSS software to determine the relationship between PM$_{2.5}$ concentrations and the meteorological parameters. The data then were analysed to determine the heavy metals and anions by applying open beaker acid digestion and ion chromatography, respectively.

2.3.1. Extraction and Analysis of Heavy Metals

Before subjecting to the extraction processes on heavy metals, the filter samples (25 mm in diameter) were prepared by cutting them into quarter equal portion using a ceramic scissor. Heavy metals were prepared by using acid digestion using the inductively coupled plasma mass spectroscopy (ICP-MS) analysis. The filter paper was soaked into digestion reagent (3 ml of concentrated nitric acid) in a Teflon vessel and shook for 10 minutes in an ultrasonic system. The vessel was heated in an oven at 180°C for 8 hours for dissolution of the particulate matter. The heated sample was filtered with a cleaned filter paper (Whatman 41, no. 1440-125) into a Teflon beaker. The liquid samples were transferred into centrifuge tube with a
total volume of about 11 ml and then heated on an open beaker at 110°C to reduce the aqueous phase to 2 ml.

Figure 1: The Penang, sampling areas, and nearby industries (Google map, 2019).

Furthermore, 1 drop of concentrated nitric acid (HNO₃) was added to each sample. The samples were subjected to analysis of selected heavy metals (Al, Fe, Na, Mg, K, Mn, Ni, Cu, Zn, V, As) using ICP-MS procedure. ICP-MS was used to examine the heavy metal concentration of Al, Fe, Na, Mg, K, Mn, Ni, Cu, Zn, V and As, with 6000cx system (ELAN DRC-e, USA). Calibration standards were prepared by diluting the stock solution with Milli-Q water or 10% nitric acid (HNO₃) with the same matrix in samples from 0.005 to 5 mg/L. In order to use, all the stock solutions were kept in refrigerated at 4°C. Nitric acids (69%), hydrogen peroxide (30%) and hydrochloric acid (36%) were obtained from Merck KGaA (Germany). National Institute of Standards and Technology (NIST) Standard reference material (SRIM) 2709a is a San Joaquin Soil (NIST, USA) and it was applied in this study to examine the behavior of acid digestion procedures. The argon gas was used in Inductively Coupled Plasma (ICP) with the purity ≥99.995% bought from Hong Kong Linde Ltd.

2.3.2. ICP-MS Analysis of Heavy Metals

ICP-MS was used to examine the heavy metal concentration of Al, Fe, Na, Mg, K, Mn, Ni, Cu, Zn, V and As. Multi-element calibration standard of ICP-MS were prepared by diluting the stock solution with Milli-Q water or 10% nitric acid (HNO₃) with the same matrix in samples from 0.005 to 5 mg/L. In order to use, all the stock solutions were kept in refrigerated at 4°C. Nitric acid (69%), hydrogen peroxide (30%) and hydrochloric acid (36%) were obtained from Merck KGaA (Germany). The argon gas was used in Inductively Coupled Plasma (ICP) with the purity ≥99.995% bought from Hong Kong Linde Ltd.

The standard solution for ICP-MS were divided into two stages; 1) 10 μg/mL mixed stock standard which contains the elements of Al, Fe, Na, Mg, K, Mn, Ni, Cu, Zn, V and As, was prepared. 1 mL of each 100 μg/mL certified standards was pipette into a clean 100 mL volumetric flask and it was diluted with ASTM Type I water containing 2% (w/w) ultrapure nitric acid until reaching 100 mL. Then, the flask was inverted and mixed well before transferring them into a clean 125 mL Teflon
bottle; 2) 100 ng/mL mixed stock standard was prepared. 1 mL of the prepared 10 μg/mL stock standard was pipette into a clean 100 mL volumetric flask and it was diluted with ASTM Type I water containing 2% (w/w) ultrapure nitric acid until reaching 100 mL. Later, the flask was inverted and mixed well before transferring them into a clean 125 mL Teflon bottle. Then, blanks of 0.1, 0.3, 0.5, 0.7, 1.0, 3.0, 5.0, 7.0 and 10.0 μg/mL calibration standards were prepared. For 100 ng/mL multi-element standard, 0, 10, 30, 50, 70, 100, 300, 500, 700 and 1000μL was pipette into ten cleaned and labelled polypropylene 15 mL centrifuge tubes. 10.00, 9.99, 9.97, 9.95, 9.93, 9.90, 9.70, 9.50, 9.30 and 9.00 mL of ASTM Type I water containing 2% (w/w) ultrapure nitric acid were added into each tube, respectively. 100 μL internal standard was pipette into each tube using an Eppendorf repeater pipette and tubes were caped and shook well.

2.3.3. Ion Chromatography (IC) Analysis

For the analysis of anions, ion chromatography method was applied. Ion chromatograph ( Dionex, DX-320, USA) with Ion Pac® AS12A (4 x 250 mm) and IonPac® CS12A (4 x 250 mm) associated with guard columns (IonPac® CG12A 4 x 50 mm and IonPac® AG12A 4 x 50 mm) were used to measure anions (Cl-, NO3-, SO42-) ions in the aqueous extract. Electrolytic self-producing suppressor components such as ASRS®300, 4 mm, for anions were consumed. The oven temperature of the LC25 chromatograph was adjusted at 35°C. The samples were injected through a 25-μL loop into an eluent. An eluent of 2.7 mM Na2CO3 (Sunnyvale, California) and 0.3 mM NaHCO3 (Sunnyvale, California) were prepared and used for detection of anions with a pump maintaining the flow rate of 1.0 mL/min.

Multi-element calibration standard of IC was prepared via consecutive additions of leading standard solution with ultrapure water (18 MΩ cm, Milli-Q System Millipore) for each ion. The calibration curves range for all ions were between 0.20 mg/L and 10 mg/L. The main stock solution was prepared from certified reference materials solutions of (1000±2) mg/L nominal ion concentration (CertiPur, Merck). All the prepared solutions were refrigerated at (4±2)°C. The calibration curve was established by plotting the peak areas for each ion against the concentration. The requirement of quality for the acceptance of the calibration function was constructed as a correlation coefficient of \( r^2 \geq 0.995 \).

Multi-element standard solution for IC was prepared by labelling 15 mL centrifuge tubes to apply for each calibration and control standards. 12 mL of the prepared standard solution was pipette to the tubes and the solution was mixed.

2.3.4. Statistical Analysis

The whole statistical assessment was carried out with the aid of SPSS software to determine significant differences between the mean concentration of insoluble materials, soluble materials, heavy metals and anions in atmosphere (Shakeri et al., 2009). The significant sources of anion and heavy metals concentration was examined using correlation analysis.

Correlation analysis was used to calculate the association between two continuous variables. The output variable was called dependent variable whereas the risk factor was called independent variables. For correlation analysis, the examined sample correlation coefficient was Pearson Product Moment correlation coefficient and the symbol for correlation coefficient was \( r \). The range of correlation coefficient was between -1 and +1 and it calculated the management and strength of the association between the two variables.

3. RESULT AND DISCUSSION

The concentration profile of PM2.5 associated together with their selected heavy metals and anions in Penang was illustrated in this section. Table 1 shows the descriptive statistics of daily mass concentration of PM2.5 in the study areas. The result revealed that PM2.5 concentrations varied in both among samples and among locations. Standard deviation calculated showed the significant difference between PM2.5 concentrations between the residential area, industrial area, and roadside. Overall, the decreasing order of PM2.5 concentrations were residential area < industrial area < roadside. This result was in line with a previous study by Beh et al. (2013) conducted in Penang as well but in island part, which reported that the PM2.5 distributions was highest due to the high number of vehicles on the road. The emission of gas from vehicles contributed to the increment of dust concentration.

Table 1: Descriptive statistics of daily mass concentration of PM2.5.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Residential (μg/m³)</th>
<th>Industrial (μg/m³)</th>
<th>Roadside (μg/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean</td>
<td>10.88</td>
<td>31.96</td>
<td>39.35</td>
</tr>
<tr>
<td>Median</td>
<td>9.73</td>
<td>26.56</td>
<td>31.43</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>6.32</td>
<td>22.07</td>
<td>22.12</td>
</tr>
<tr>
<td>Maximum</td>
<td>28.47</td>
<td>80.56</td>
<td>79.51</td>
</tr>
<tr>
<td>Minimum</td>
<td>4.86</td>
<td>9.03</td>
<td>18.06</td>
</tr>
<tr>
<td>No. of samples</td>
<td>12</td>
<td>12</td>
<td>12</td>
</tr>
</tbody>
</table>

According to Dahari et al. (2020), due to the industrialization, use of vehicles, and expansion of suburban areas into proximity with industrial areas, the particle pollution in ambient environment is increasing.

Table 2 shows the result of the correlation between PM2.5 concentration and meteorological parameters in the sampling areas comprised of residential area, industrial area, and roadside. This correlation was based on the daily concentration of PM2.5 in the areas. The
result indicated that negative correlation \((r = -0.207)\) was found between \(\text{PM}_{2.5}\) and precipitation. The similar result was reported by Owoade et al. (2012). This is due to the washout process as the rain exhibit wet deposition effect on particulate matter, thus, reduced the mass concentration of the atmospheric particulate. Precipitation also had effectively removed the atmospheric particulate matter, especially due to the smaller size (Wang and Ogawa, 2015).

**Table 2**: Correlation between \(\text{PM}_{2.5}\) and the meteorological parameters in the study areas.

<table>
<thead>
<tr>
<th>Correlation</th>
<th>R</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{PM}_{2.5}) with precipitation</td>
<td>-0.207</td>
<td>36</td>
</tr>
<tr>
<td>(\text{PM}_{2.5}) with wind speed</td>
<td>0.081</td>
<td>36</td>
</tr>
<tr>
<td>(\text{PM}_{2.5}) with temperature</td>
<td>0.155</td>
<td>24</td>
</tr>
<tr>
<td>(\text{PM}_{2.5}) with humidity</td>
<td>-0.343</td>
<td>24</td>
</tr>
<tr>
<td>(\text{PM}_{2.5}) with air pressure</td>
<td>0.476*</td>
<td>24</td>
</tr>
</tbody>
</table>

*Correlation is significant at the 0.05 level (2-tailed).

Then, positive correlation coefficient with the value of 0.081 was found between \(\text{PM}_{2.5}\) and wind speed. Wind speed plays a major role in cleansing atmosphere of fine particulates as the pollutant transportation and dispersion agent (Dahari et al., 2020). However, wind speed in study areas affected the turbulence near the ground ineffectively. It means the higher the wind speed, the higher the distribution of \(\text{PM}_{2.5}\) concentrations in the area. According to Wang and Ogawa (2015), when the wind speed is high enough, it can transport large quantities of pollutants from far away. This result is parallel to the result obtained in Penang, where the dominant wind speed ranges between 6 to 9 m/s. Apart from wind speed, wind direction also a major factor that influenced fine particulate matter by transporting from different neighbouring regions to the sampling location. However, since wind direction only indication of where the direction of the wind came from and not a linear variable, it is not considered in the analysis.

\(\text{PM}_{2.5}\) exhibits strong positive correlation with temperature. This result is in an agreement with other studies where fine particles being favoured of the transformations which took place in the atmosphere with the presence of solar radiation reached their peak values during the highest temperature days (Pateraki et al., 2012). High temperatures were clearly conducive to intense convection. Atmospheric \(\text{PM}_{2.5}\) was transported quickly and effectively, allowing its accelerated dispersion. Therefore, the local mass concentrations were decreased. Conversely, low temperatures and the temperature inversion layer caused by radiative cooling weakened convections. In these circumstances, \(\text{PM}_{2.5}\) remained suspended under the inversion layer, leading to higher concentrations (Li et al., 2015).

\(\text{PM}_{2.5}\) yielded a negative correlation with relative humidity as well as statistically significant at 0.05 confidence level. Due to the hygroscopic growth, \(\text{PM}_{2.5}\) concentration decreases when humidity is low. However, when the humidity is high enough, the particles grew heavy to stay in the air. Hence, dry deposition occurred and caused the particles fell to the ground. Consequently, particle numbers reduced, and concentration decreased (Wang and Ogawa, 2015).

Mass concentration displays a strong positive relation with air pressure. The result is corresponding to the result reported by Li et al. (2015) suggesting the surface experiences a convergence up draft when controlled by low pressure and the up draft promoted the dispersion of \(\text{PM}_{2.5}\) from the ground up into the air. Thus, mass concentrations at the sampling point are subsequently reduce. Comparatively, when there was high pressure, the down draft restrained the upward movement of mass concentration caused an accumulation of particles (Li et al., 2015).

**3.1. Heavy metals and anion ions concentration levels**

The concentration of heavy metals such as Al, Fe, Na, Mg, K, Mn, Ni, Cu, Zn, V and As as well as the target anions such as Cl\(^-\), NO\(_3^-\) and SO\(_4^{2-}\) determined in \(\text{PM}_{2.5}\) samples collected from industrial, residential and roadside is presented in Tables 3. The result for heavy metal concentration reveals that K, Na, Zn and Mg were dominant at the roadside area and industrial area of Mainland, Penang. Mean concentration of K at the roadside, industrial area and residential areas are 354.82 ng/m\(^3\), 324.66 ng/m\(^3\), and 216.80 ng/m\(^3\), respectively. Na concentrations value are 159.24 ng/m\(^3\), 143.32 ng/m\(^3\) and 137.96 ng/m\(^3\) at the above mentioned three areas, respectively.

High concentration of K and Na were found at three sampling areas. The highest number of automobile emissions, sea spray (on-shore winds forms ample sea water droplets with marine atmosphere) and coal combustion from nearby construction site may influence the highest concentration of K. K and Na are metals of crustal elements which may be came from doubled soil and road dust resuspension during the respective sampling days and the result of Asian dust which comes from soils in the dry desert and upland areas in spring also influence for these metals. This statement is supported by Ec-Ling et al. (2015), saying that the heavy metal such as K is subjected to be the highest emission in \(\text{PM}_{2.5}\) of the total measured elemental concentration Zn, Mg, Fe, Al, Ni, Cu, A\(_s\), Mn and V were almost similar throughout the sampling period at roadside and industrial area. Roadside is situated nearby industrial area where the emissions come from the burning of fossil fuels, coal combustion, road dusts, traffic pressure and different industry activities travel among those areas. The overall decreasing order of metal concentrations were K> Na> Zn> Mg> Fe> Al> Ni> Cu> A\(_s\)> V> Mn, K> Na> As> Fe> Al> Ni> Cu> A\(_s\)> V> Mn, K> Na>
The study concluded that the meteorological parameters play a major role in day-to-day variations of the mass concentration of PM$_{2.5}$ together with its chemical composition such as heavy metals and anion ions. The locations also highly influenced the distribution of PM$_{2.5}$. The highest mean concentration was found in roadside area with 39.35 μg/m$^3$, followed by industrial area (31.96 μg/m$^3$) and lastly residential area (10.88 μg/m$^3$). Hence, the sources of PM$_{2.5}$ were most likely influenced its anthropogenic sources in the respective areas. Meanwhile, result of Pearson correlation suggested the PM$_{2.5}$ concentration in the study areas less impacted by the meteorological parameters as the only significant correlation coefficient obtained from PM$_{2.5}$ and air pressure with 0.476. SO$_4^{2-}$, K and Na were found as dominant chemical in the PM$_{2.5}$ of Penang. The mean concentration of SO$_4^{2-}$, K and Na were 4.42 μg/m$^3$, 354.82 ng/m$^3$, 159.24 ng/m$^3$ at roadside. The mean concentration of SO$_4^{2-}$, K and Na were 4.88 μg/m$^3$, 324.66 ng/m$^3$ and 143.32 ng/m$^3$ at industrial area, roadside, and residential area, respectively.

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