

Spatial and Temporal Analysis of Particulate Matter (PM₁₀) in Urban-Industrial Environment during Episodic Haze Events in Malaysia

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Abstract

Haze episode in Malaysia typically takes place during the dry monsoon season. As a result, high concentration of atmospheric particles was recorded primarily brought by transboundary air pollution from the neighbour country. Therefore, this study aims to evaluate and compare the level of particulate matter (PM_{10}) at urban-industrial areas during the episodic haze episodes in Malaysia. Hourly PM_{10} concentration with the concentration of gaseous air pollutants such as NO_x, NO₂, SO₂, CO and O₃ and meteorological parameters (relative humidity, temperature, wind speed) at urban-industrial areas namely Shah Alam (Selangor), Nilai (Negeri Sembilan), Bukit Rambai (Melaka) and Larkin (Johor), during the haze episode in 1997, 2005, 2013 and 2015 were used for analysis. In this study, spatio-temporal and correlation analysis were used to provide an overview of the distribution pattern and examine the relationships between the gaseous air pollutants and meteorological parameters with PM₁₀ concentration. From the descriptive statistics, it was observed that PM₁₀ level for all study areas were skewed to the right (> +1) indicating occurrences of extreme events. A significant peak of PM₁₀ concentration for each year of haze events were observed to be started in June or during the southwest monsoon to the inter monsoon in October. The occurrence, duration and impact of 1997 haze was detected to be identical to the 2015 haze event that reached its peak in October. From the correlation analysis, PM_{10} concentration were strongly correlated to the CO concentration (r > 0.5) during High Particulate Event (HPE). Very weak relationship of PM₁₀ level with meteorological parameters (r < 0.3) were observed. Interestingly, O₃ level shows very strong correlation with the meteorological parameters during HPE. The findings provide comprehensive evaluation on PM₁₀ level during the historic haze episodes, thus can help the authorities in developing policies and guidelines to effectively monitor and reduce the negative impact of haze events.

Keywords: Air quality; Air quality modeling; Haze; Particulate Matter (PM₁₀); Pearson correlation

1. Introduction

Extremely high concentration of particulate matter especially with an aerodynamic diameter $\leq 10 \mu m (PM_{10})$ in ambient air usually occurred during haze episodes or also known as high particulate event (HPE). These severe episodes often occurs when the concentrations was far exceeded the Malaysian Ambient Air Quality Standard (MAAQS) for PM_{10} concentration which was 100 µg/m³ for 24-hour average. Haze in Malaysia is not a new phenomenon as it was first recorded back in the year 1982 (Tonoto, 2017). Since then, several haze episodes have been recorded. These severe episodes occurred in 1997, 2005 and 2015 (Latif et al., 2018).

The worst haze episode occurred in 1997 when nearly the entire country was engulfed by thick smog for almost six months (Othman et al., 2014). It was recorded that a 24-h average concentration of PM₁₀ near to Kuala Lumpur city center has exceeded 300 µg/m³ (Abas et al., 2004). The Air Pollution Index (API) recorded during 1997 had also exceeded a very unhealthy level which was 350 in September (Mahmud, 2009). Another severe HPE occurred during 2005 which is mainly on the central west coast of the Peninsular Malaysia (Sahani et al. 2014). Severe smoke haze episodes occurred in 11 August in two areas namely Pelabuhan Klang and Kuala Selangor with PM₁₀ concentration exceeded 500 μ g/m³ and then shifted to the northern states by 13 August 2005 (DoE, 2013).

In 2013, HPE occurs mainly during June and July. Muar, Johor recorded the highest peak that was 746 µg/m³ which was almost 2.5 times the lower end of the hazardous level, thus resulting in the declaration of a state of emergency in Muar (Show and Chang, 2016). The latest and the worst HPE that had been recorded in Malaysia's history was in 2015 which was lasted for two month (Latif et al., 2018). According to Department of Environment (DoE, 2016) report, all schools in the states of Putrajaya, Kuala Lumpur, Selangor, Negeri Sembilan and Melaka were closed due to the API reading reaching to the category of unhealthy and very unhealthy air. As Malaysia experiences recurring haze events, the causes of these HPE varies in many ways. Internally, the causes of these HPE was generated roughly from local anthropogenic sources originating from various sources such as industrial activities and vehicles usage (Khan *et al.*, 2020; Latif *et al.*, 2018). It became worse when the smog from forest fires and slash-and-burn land clearing in Sumatra permeates the Malacca straits and reached Malaysia (Sue Wen *et al.*, 2016). Transboundary pollution may originate by Indonesia's deforestation and forest fires, but unpredictably anomalous weather (wind direction and dry season) may influence and exacerbated the severity of the HPE (Ku Yusof *et al.*, 2017).

Haze can cause mortality and serious illness such as respiratory and cardiovascular related diseases. Koplitz et al., (2016) reported that during the HPE in 2015, 100,300 deaths were recorded across Indonesia, Malaysia and Singapore. It was also observed that the number of diagnostics of lung cancer patients have been dramatically increased during haze period due to their cancer with acute symptoms using the hospital admission data from 2010 to 2015 (Hassan et al., 2017). Aside from the impact of these HPE event towards health, economic sector in Malaysia also suffered a severe loss. It is stated that the estimated economic loss of the slash and burn practices that caused by the Southeast Asia forest fires in 1997 was USD 20.1 billion (Varma, 2003).

Meteorological conditions often associated as a major influence towards PM₁₀ concentration. Several studies indicated that PM₁₀ concentration show positive correlation with ambient temperature (Noor et al., 2015; Siti et al., 2015). It was stated that the increase in temperature usually rises the quantity of biomass burning and the evaporation of materials causing the increase of PM₁₀ concentration. In contrast with ambient temperature, PM₁₀ has a reverse relationship with relative humidity and wind speed (Alifa et al., 2020; Payus et al., 2013). Relative humidity is commonly affected by the number of rain occasions which through wash-out processes of the atmospheric aerosols (Afzali et al., 2014; Gvozdić et al., 2009) and increase in wind speed would cause PM₁₀ to dilute by dispersion (Akpinar et al., 2008)

in which resulted to reduction of concentration of pollutant in the air.

Currently, several studies used statistical methods to assess the status of air quality in Malaysia. Alifa et al. (2020) investigated the influence of urban and wildfire emissions, as well as the key meteorological variables on the observed PM_{10} patterns through a time series analysis and explored on a seasonal basis using multiple linear regression. Othman et al. (2021) also studied the partial distribution of particulate matter during southwest monsoon in peninsular Malaysia using one-way ANOVA with Turkey post-hoc test and spatial interpolation inverse distance weighting (IDW) technique to quantify the spatial and temporal concentrations of the organic components for holistic PM_{10} chemical compositions characterization. These researches aim to provide both baseline and a reference for future studies as well as pollution mitigation strategies and climate change (Alifa et al., 2020).

Therefore, this study focus on evaluating and comparing the characteristics and behavior of PM_{10} level at the four areas in the west coast of peninsular Malaysia during the historic HPE in Malaysia. The results from this study may be useful as it provide scientific information during episodic HPE, thus strategic development on the policies involving local and international authorities can be directed. This study supported SDG2030 in Goal 11 (Sustainable Cities and Communities) which is aimed to reduce the adverse per capita environmental impact of cities, by paying special attention to air quality (annual mean levels of fine particulate matter).

2. Materials and Methods

2.1 Study area

In this study, 4 stations that represent urban-industrial environment located in peninsular Malaysia namely Larkin (Johor), Bukit Rambai (Melaka), Nilai (Negeri Sembilan) and Shah Alam (Selangor) were chosen. Urbanisation is closely linked with modernisation, industrialization and the sociological process of rationalisation (Bhatta & Bandyopadhyay, 2010). Figure 1 and Table 1 describe the location, population and background of each of the monitoring stations. Larkin, Bukit Rambai and Nilai are located at the southern region of peninsular Malaysia whereas Shah Alam is sited at the centre. All of these stations were prone to the transboundary smoke from the Sumatera regions as these stations resides at the west coast of peninsular Malaysia.



Figure 1. Location of the study areas that are located in the peninsular Malaysia

| Monitoring Station | Latitude (N) | Longitude (E) | Population (Department of Statistics Malaysia, 2021) | Background of Study Area | | |
|---------------------------|--------------|---------------|---|-----------------------------|--|--|
| Shah Alam, Selangor | 03°06.286' | 101°33.367' | 617,149 | Urban Industrial | | |
| Nilai, Negeri Sembilan | 02°49.246' | 101°48.877' | 692,283 | Urban Industrial | | |
| Bukit Rambai, Melaka | 02°12.789' | 102°14.364' | 597,135 | Urban Industrial | | |
| Larkin, Johor | 01°28.225' | 103°53.637' | 1,711,191 | Urban Industrial | | |

Table 1. Details of the study areas

* Urban-Industrial indicates the areas due to large population, hence a lot of residential and commercial areas can be found near by

2.2 Data Collection

The air quality data were collected from the Air Quality Division of the Department of Environment (DoE), Malaysia. The equipment used by Alam Sekitar Malaysia Sdn. Bhd (ASMA) to monitor the air quality data are from Teledyne Technologies Inc. USA, and Met One Instrument Inc. USA. Table 2 lists the details of equipment used by ASMA. The data that were obtained from the DoE, Malaysia was commonly subjected to standard quality control processes and quality assurance procedures (Mohammed et al., 2013). The procedures that were used in the monitoring stations followed the standards outline States Environmental Protection Agency (USEPA) (Latif et al., 2014).

Continuous hourly data of air pollutants and meteorological parameters in the year that Malaysia experienced historic HPE (1997, 2005, 2013 and 2015) were chosen in this study. Table 3 shows the air pollutants and weather parameters that were obtained from DOE, Malaysia.

2.2.1 Data Pre-treatment

The missing observation of PM₁₀, gases pollutants and meteorological parameters

were first fill-in before the analysis were done. These missing data was treated using Linear Interpolation (LI) method using Statistical Package for the Social Sciences (SPSS) Software Version 26 (Zakaria and Noor, 2018).

2.2.2 Data Analysis

Descriptive analysis was conducted by using SPSS version 26 to analyse the characteristic of the dataset. Measure of central tendency was used to investigate the centre or the middle of the air pollution dataset which includes mean, median and mode. Furthermore, measure of dispersion was used to study the dispersion of values around the central tendency which includes standard deviation, variance, skewness, maximum and minimum. Then, spatial analysis was performed using Ordinary Kringing Interpolation Method (OKM) using ArcGIS software version 10.5 to analyze continuous distribution of PM₁₀ concentration among the stations (De Iaco et al., 2005). OKM is widely applied to study the spatiotemporal distribution of air quality, and the corresponding model has the best superiority in cross validation (Fan et al., 2020).

| Table 2. Details of equipment for air pollutants monitoring in Malaysia (Standar | d Operating |
|--|-------------|
| Procedures for Continuous Air Quality Monitoring, 2007) | |

| Air Pollutant | Instrument | Analyzer | Detection limit |
|-------------------------------------|--------------|--------------------------|---------------------------------|
| PM ₁₀ | Met One | BAM-1020 Beta | < 4.8 µg/m ³ and $<$ |
| | Instrument, | Attenuation Mas | 1.0 μg/m ³ for 1-h |
| | Inc. USA | Monitor | and 24-h |
| SO ₂ | | UV fluorescence | 0.5% precision and |
| | | method | the lowest detection |
| | | | of 0.4 ppb |
| СО | | Non-dispersive, infrared | 0.5% precision and |
| | Teledyne | absorption (Beer | the lowest detection |
| | Technologies | Lambert) method | of 0.04 ppm |
| O ₃ | Inc., USA | UV absorption (Beer | 0.5% precision |
| | | Lambert) method | detection limit of 0.4 |
| | | | ppb |
| Non-methane | | Aflame-ionization | Measurement |
| Hydrocarbon | | detector (FID) | accuracy of 1% |
| NO/NO ₂ /NO _X | | Analyzer model 200A | - |
| | | with microprocessor | |
| | | technologies | |

Table 3. Air pollutants and meteorological parameters

| Air Quality & Weather Parameters | Abbreviation | Unit | |
|-------------------------------------|-----------------|-------------|--|
| Particulate Matter | PM_{10} | $\mu g/m^3$ | |
| Nitrogen oxides | NO _x | ppm | |
| Sulphur dioxides | SO_2 | ppm | |
| Surface ozone | O ₃ | ppm | |
| Nitrogen dioxides | NO_2 | ppm | |
| Carbon monoxide | СО | ppm | |
| Temperature | Т | °C | |
| Windspeed | WS | km/h | |
| Relative humidity | RH | % | |

Lastly, Pearson's correlation coefficient was calculated to evaluate the linear relationship of PM_{10} concentration and other gaseous pollutants and meteorological parameters during HPE. The degree of the correlation can be identify by the calculated Pearson correlation (r) value. Correlation ranges from -1.0 to 1.0 which can be considered as negatively correlated or positively correlated. If the correlation coefficient (r) is higher than 0.6, it was declared as high correlation; moderately correlation with the r-value between 0.3 - 0.6 and; weak correlated with the r-value smaller than 0.3 (Gogtay and Thatte, 2017). The formula for the correlation coefficient was given as follows (Awang *et al.*, 2016):

$$r = \frac{\sum_{i=1}^{n} (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^{n} (x_i - \bar{x})^2} \sqrt{\sum_{i=1}^{n} (y_i - \bar{y})^2}}$$

where x_i and y_i are the ith sample values of PM₁₀ measurements and other parameters respectively; \overline{x} and \overline{y} are the mean value of PM₁₀ measurement and other parameters correspondingly.

2.2.3 Trajectory Analysis

A trajectory analysis using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) was conducted to determine the origin of the air masses backward trajectories for 120-hours (4 days) during HPE. The model used in this study is the NOAA (HYSPLIT-4). The model calculation method is a hybrid between the Lagrangian approach, using a moving frame of reference for the advection and diffusion calculations as the trajectories or air parcels move from their initial location, and the Eulerian methodology, which uses a fixed three-dimensional grid as a frame of reference to compute pollutant air concentrations (Stein et al., 2015).

3. Results and Discussion

3.1 Variation of PM_{10} during HPE

Table 4 shows the descriptive statistics of hourly PM₁₀ concentration in the study areas. Overall, all study areas had more than 80% valid measurement of PM₁₀. Bukit Rambai recorded the highest mean value among all stations during 1997, meanwhile, Nilai shows highest mean value in 2013 and 2015, whereas Shah Alam has the highest mean value in 2005. Overall, the mean value observed in all stations during all respective years were higher than the median value. It indicates that the distribution of PM₁₀ concentration was positively skewed hence higher possibility of PM₁₀ measurement records exceeded the permissible value of Recommended Malaysia Ambient Air Quality Guideline (RMAAQG) which was 150 µg/m³ (for 24-h average). It can be confirmed with the skewness value where all areas specify the value that was greater than +1, which show that the data was highly skewed to the right. PM₁₀ concentration for Bukit Rambai, Nilai and Shah Alam were recorded having the highest standard deviation in 1997 with 61.59µg/m³, 64.24µg/m³ and 62.88µg/m³ respectively due to high variability in dataset distribution. The highest maximum value of PM10 level was recorded at Shah Alam in 2005 (587 µg/m³) followed by Bukit Rambai (2013) and Nilai (1997). In general, all the maximum values in the respective year of study shows high value of maximum hourly PM₁₀ concentration which exceeded the RMAAQG.

| | | | Bukit F | Rambai | | | | | |
|---|----------|-------|---------|--------|--------|-------|--------|-------|-------|
| | | 1997 | 2005 | 2013 | 2015 | 1997 | 2005 | 2013 | 2015 |
| Ν | Valid | 8337 | 7427 | 8163 | 8759 | 8436 | 8757 | 8644 | 8620 |
| | Missing* | 423 | 1333 | 597 | 1 | 324 | 3 | 116 | 140 |
| | | (4.8) | (15.2) | (6.8) | (0.01) | (3.7) | (0.03) | (1.3) | (1.6) |

Table 4. The descriptive statistics of PM_{10} concentration in 1997, 2005, 2013 and 2015

| Ν | Valid | 8337 | 7427 | 8163 | 8759 | 8436 | 8757 | 8644 | 8620 | | | |
|-----|---|---------|--------|---------|---------|-----------|---------|--------|---------|--|--|--|
| | Missing* | 423 | 1333 | 597 | 1 | 324 | 3 | 116 | 140 | | | |
| | | (4.8) | (15.2) | (6.8) | (0.01) | (3.7) | (0.03) | (1.3) | (1.6) | | | |
| Me | an | 71.70 | 57.13 | 57.24 | 69.69 | 64.16 | 63.18 | 58.17 | 69.90 | | | |
| Me | edian | 46.00 | 52.00 | 50.00 | 58.00 | 40.00 | 56.00 | 52.00 | 58.00 | | | |
| Mo | ode | 35.00 | 54.00 | 54.00 | 52.00 | 30.00 | 52.00 | 54.00 | 58.00 | | | |
| Sto | l. Deviation | 61.59 | 20.05 | 43.75 | 41.47 | 64.24 | 33.57 | 29.74 | 44.02 | | | |
| Va | riance | 3793.23 | 401.92 | 1913.70 | 1720.00 | 4127.38 | 1127.24 | 884.33 | 1938.00 | | | |
| Sk | ewness | 2.45 | 1.99 | 5.58 | 2.70 | 3.24 | 4.14 | 3.67 | 2.63 | | | |
| Mi | nimum | 13.00 | 18.00 | 18.00 | 24.00 | 13.00 | 19.00 | 17.00 | 13.00 | | | |
| Ma | aximum | 415.00 | 178.00 | 515.00 | 338.00 | 510.00 | 330.00 | 304.00 | 353.00 | | | |
| | | | La | rkin | | Shah Alam | | | | | | |
| Ν | Valid | 8308 | 8622 | 8692 | 7928 | 4580 | 8744 | 8608 | 8639 | | | |
| | Missing* | 451 | 138 | 68 | 832 | 556 | 16 | 152 | 121 | | | |
| | | (5.1) | (1.6) | (0.8) | (9.5) | | (0.2) | (1.7) | (1.4) | | | |
| Me | ean | 36.35 | 35.55 | 44.15 | 67.34 | 66.76 | 64.48 | 46.39 | 60.27 | | | |
| Me | edian | 25.00 | 34.00 | 39.00 | 52.00 | 42.00 | 54.00 | 42.00 | 51.00 | | | |
| Mo | ode | 20.00 | 31.00 | 36.00 | 39.00 | 19.00 | 52.00 | 52.00 | 49.00 | | | |
| Sto | l. Deviation | 31.99 | 10.87 | 31.99 | 56.84 | 62.88 | 49.32 | 30.04 | 38.60 | | | |
| Va | riance | 1023.07 | 118.09 | 1023.14 | 3231.25 | 3953.95 | 2431.99 | 902.68 | 1489.81 | | | |
| Sk | ewness | 2.62 | 1.34 | 5.84 | 3.02 | 1.98 | 5.83 | 6.29 | 3.28 | | | |
| Mi | nimum | 8.00 | 14.00 | 10.00 | 5.00 | 10.00 | 14.00 | 12.00 | 5.00 | | | |
| Ma | ximum | 208.00 | 108.00 | 368.00 | 489.00 | 368.00 | 587.00 | 421.00 | 455.00 | | | |
| *T | The parameter of missing data is given in the parameters in | | | | | | | | | | | |

The percent of missing data is given in the parenthesis.

Figure 2 depicts the comparison of daily PM_{10} level in 1997, 2005, 2013 and 2015 for all study areas. Overall, a significant peak indicating HPE for was observed to be started in June during the southwest monsoon to the inter monsoon in October. In 1997, PM_{10} level started to rise in early September and reached the peak at the end of September and October. Bukit Rambai and Shah Alam were monitored to have almost the same intensity of PM_{10} level during the peak, however Nilai recorded the highest concentration of almost 500 µg/m³.

The least concentration of PM_{10} during the peak was recorded in Larkin with the concentration of 200 µg/m³ that was slightly high from the value of RMAAQG (150 µg/m³). Since there was a large-scale air quality disaster that occurred during the second half of 1997, it was expected that all stations experienced HPE. It can be evident from the peak in September and October before it is weakening by November, when the delayed monsoonal rain extinguished the fires and improved air quality within the region (Jim, 1999).



Figure 2. Daily time-series plot of PM₁₀ concentration (a) 1997, (b) 2005, (c) 2013 and (d) 2015. The red line indicate the recommended daily permissible level of PM₁₀ level according to RMAAQG that was 150 μg/m³

During 2005, PM₁₀ concentration at Shah Alam exceeded the RMAAQG value in early March and April. The daily peak of PM_{10} level was observed in August in which the highest average daily PM₁₀ concentration was recorded at Shah Alam (537.58 µg/m³) and followed by Nilai (301.83 µg/m³). According to the Department of Environment (2016), the severe HPE occurs on early August and affected mostly on the central, eastern and northern parts of peninsular Malaysia. Since Shah Alam and Nilai are located at the central part of peninsular Malaysia, it was expected that the monitoring station has the highest average daily PM₁₀ records, meanwhile, Larkin and Bukit Rambai were unaffected.

In 2013, PM₁₀ level started to significantly rise up in in the end of June and reached its peak on early July. Different from other years of HPE that have been discussed earlier, all study areas were badly affected with the maximum concentration of PM₁₀ in Bukit Rambai reaching > 400 μ g/m³. Whereas in 2015, it can be seen that the variation of PM₁₀ level during the HPE was similar to the 1997. All areas were affected with the highest PM₁₀ level was observed in Shah Alam. Furthermore, HPE in 2015 were the longest recorded HPE that lasted for about 2 months and it was stated as the worst haze period in Malaysia's history after 1997 (Latif *et al.*, 2018).

From Figure 2, the HPE often occur starting around June until October and the peak of the event for all areas seem to be during September and October. The transboundary air pollution caused by the fire burning on the Indonesia island of Sumatras was the major attribute to the HPE, where the strong wind brings the fumes across the Melaka Strait during the dry monsoon season. In addition, the peak of PM_{10} concentration levels were mostly detected during June-September, which is during southwest monsoon where low-level winds were associated with long-range transportation of pollutants (Noor *et al.*, 2015). These fine particles are able to be transported across borders from Sumatra and Kalimantan northward to peninsular Malaysia due to the regional wind direction (Amil *et al.*, 2016).

Table 5 shows the total hours and days of PM_{10} exceedances according to RMAAQG that was > 150 µg/m³. Bukit Rambai station recorded the highest number of hours exceeding the RMAAQG in 1997, 2013 and 2015 with the total of 795, 234 and 608 hours in the respective years. In Figure 2 (a), Nilai shows the highest peak of PM₁₀ concentration among all stations in 1997 which can be evidenced in Table 5. Furthermore, Nilai monitoring station recorded the highest total hours in 2005 with 249 hours followed by Shah Alam with 243 hours. Larkin recorded the least numbers of exceedances with no exceedances in 2005.

In order to get clearer view on the monthlybased exceedances, Table 6 describes the exceedance according to year and months. The exceedances of total day of PM₁₀ level exceeded the RMAAQG limit was recorded in September and October. The reoccurrence of trans-boundary haze pollution causes the deterioration of air quality and usually occurs during the summer monsoon where the number of hotspots recorded in Sumatra and Kalimantan, Indonesia escalates (Raffee et al., 2019). Southwest monsoon largely influenced phases of the El Nino phenomenon. These dry conditions coupled with the warm temperatures associated with El Nino creates an extremely favorable and conducive environment for large-scale outbreaks in Sumatra and Kalimantan (Latif et al., 2018).

Table 5. Summary of total hours and days of PM_{10} concentration exceeding the RMAAQG (> 150 µg/m³) during 1997, 2005, 2013 and 2015

| | 1997 | | 2005 | | 2013 | | 2015 | | |
|--------------|------|----|------|----|------|----|------|----|--|
| Station | TH | TD | TH | TD | TH | TD | TH | TD | |
| Bukit Rambai | 795 | 32 | 41 | 7 | 234 | 10 | 608 | 22 | |
| Larkin | 174 | 13 | 0 | 0 | 155 | 7 | 332 | 45 | |
| Nilai | 712 | 40 | 249 | 14 | 140 | 8 | 485 | 29 | |
| Shah Alam | 489 | 27 | 243 | 15 | 77 | 4 | 521 | 54 | |

*Total Hours (TH) - Number of hours exceeding the 1-h-PM₁₀ RMAAQG of 150 μ g m-3 **Total Days (TD) - Days with at least one of hourly PM₁₀ measurement exceeding RMAAQG

| Station | Year | Jan | Feb | Mar | Apr | May | Jun | Jul | Aug | Sept | Oct | Nov | Dec | Total |
|-----------|-------|-----|-----|-----|-----|-----|-----|-----|-----|------|-----|-----|-----|-------|
| | 1997 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 10 | 23 | 12 | 3 | 0 | |
| Dulat | 2005 | 0 | 2 | 1 | 0 | 0 | 1 | 1 | 2 | 0 | 0 | 0 | 0 | |
| Rambai | 2013 | 0 | 0 | 0 | 0 | 0 | 11 | 1 | 0 | 0 | 0 | 0 | 0 | |
| Tunnour | 2015 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 17 | 18 | 0 | 0 | |
| | Total | 0 | 2 | 1 | 0 | 0 | 12 | 3 | 12 | 40 | 30 | 3 | 0 | 103 |
| | 1997 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 9 | 4 | 0 | 0 | |
| | 2005 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | |
| Larkin | 2013 | 0 | 0 | 0 | 0 | 0 | 7 | 0 | 0 | 0 | 0 | 0 | 0 | |
| | 2015 | 1 | 1 | 0 | 0 | 2 | 0 | 1 | 0 | 21 | 17 | 1 | 1 | |
| | Total | 1 | 1 | 0 | 0 | 2 | 7 | 1 | 0 | 30 | 21 | 1 | 1 | 65 |
| | 1997 | 0 | 0 | 0 | 0 | 0 | 0 | 4 | 4 | 24 | 6 | 2 | 0 | |
| | 2005 | 0 | 3 | 2 | 0 | 0 | 0 | 0 | 9 | 0 | 0 | 0 | 0 | |
| Nilai | 2013 | 0 | 0 | 0 | 0 | 0 | 8 | 0 | 0 | 0 | 0 | 0 | 0 | |
| | 2015 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 16 | 13 | 0 | 0 | |
| | Total | 0 | 3 | 2 | 0 | 0 | 8 | 4 | 13 | 40 | 19 | 2 | 0 | 91 |
| | 1997 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 2 | 21 | 3 | 1 | 0 | |
| | 2005 | 0 | 5 | 2 | 0 | 0 | 0 | 0 | 8 | 0 | 0 | 0 | 0 | |
| Shah Alam | 2013 | 0 | 0 | 0 | 0 | 0 | 4 | 0 | 0 | 0 | 0 | 0 | 0 | |
| | 2015 | 0 | 0 | 2 | 0 | 1 | 3 | 3 | 3 | 22 | 20 | 0 | 0 | |
| | Total | 0 | 5 | 4 | 0 | 1 | 7 | 3 | 13 | 43 | 23 | 1 | 0 | 100 |

Table 6. Summary of total days according to month of PM_{10} concentration exceeding RMAAQG during 1997, 2005, 2013 and 2015

From previous analysis, it can be determine that seasonal variation of PM₁₀ level seem much related to southwest monsoon. Figure 3 presents contour plot of average monthly PM₁₀ concentration during the southwest monsoon (June-September) and intermonsoon (October). Overall, high monthly concentration was observed during the end of southwest monsoon especially in August and September whereas, the concentration drops significantly in November when the northeast monsoon started. The most affected areas were Bukit Rambai, Nilai and Shah Alam whereas Larkin seems to have moderate to less effect of HPE. The three most affected areas were situated directly opposite to Sumatera, where most of the transboundary air pollution episodes in SEA originated from there. A study by Elhadi et al., (2018) revealed that the southwest monsoon wind from Sumatra reaches the central area of peninsular Malaysia within 48-h during southwest monsoon season prevailing every year. In June and July, Bukit Rambai was observed to experience the worst HPE compared to the other locations while Shah Alam and Nilai were detected to have a greater effects of HPE during September and October. Larkin may not be the area that can be classified as badly affected by HPE, however, the monthly

average of PM₁₀ level still exceeded the permissible value of RMAAQG.

Figure 4 shows the backward trajectories that were used to calculate the 120-hours (four days) at the height of 500m above ground level (AGL). Larkin was excluded from this analysis since it has no to minimal effects of HPE as discussed earlier. From Figure 3 (a), the trajectory during haze in August 2005 showed that the air masses clearly originated from Sumatra, arriving the study area from southwest direction. In addition, figure 4 (b) also show that air masses in June 2013 also originated from Sumatra which specifically the north Sumatra. It was proven by a study conducted by Show and Chang (2016) that as many as 676 fire activities were recorded in Sumatra on 19 June 2013 which counted as prominent peak hotspot. During this season, southwesterly wind blowing from Sumatra to Malaysia and bring along thick smoke blanketing Singapore and part of Malaysia for weeks (Noor et al., 2015). Figure 4 (c) illustrates the backward trajectory during early October 2015 that showing the air masses travels from Kalimantan region. A study by Khan et al. (2020) reported that the emission of CO flux in Kalimantan was about 6-7 times higher in strength than at Sumatra during the fire events of 2015.



Figure 3. Mapping of seasonal PM_{10} concentration distribution at the study areas



Figure 4. Backward trajectories during HPE in (a) 2005; (b) 2013 and; (c) 2015 at Bukit Rambai, Shah Alam and Nilai

In general, peninsular Malaysia receive strong winds that have travelled across the islands of Indonesia which carrying transboundary smoke from peat and forest fires (Urbančok *et al.*, 2017) either from Sumatra or Kalimantan. It was estimated to contribute to the PM₁₀ pollution in peninsular Malaysia during the southwest monsoon (Othman *et al.*, 2021).

3.2 Correlation Analysis

The relationship between PM_{10} concentration with the meteorological parameters and gaseous pollutants is given in Figure 5. Overall, all study areas shows significant correlation of PM₁₀ concentration with CO. In Bukit Rambai, strong correlation was observed between PM₁₀ and CO concentrations with r-value of 0.67; 0.54 in Larkin and 0.64 in Nilai; and the highest r-values was calculated in Shah Alam that was 0.68. All locations in this study are classified as industrial and or urban-industrial area. CO which mainly released by motor vehicle and machinery that used diesel fuel seem to have the strongest correlation with PM_{10} concentration. This may contribute to this positive correlation results. Furthermore, the seasonal fires from Indonesia can also be the main contribution during the HPE. It was reported by Huijnen et al. (2016) that the seasonal fires was greatly inflated during El Niño and drought which causes large amounts of terrestrially-stored carbon into

the atmosphere, primarily in the form of CO_2 , CO and CH_4 .

Moderate positive correlation of PM₁₀ level was detected with SO₂ concentration in Bukit Rambai (r = 0.33) and negative correlation was observed in Larkin (r = -0.32). Bukit Rambai is situated next to Ayer Keroh industrial area and this area is also surrounded by a few coal-fired power plant. High concentration of SO₂ in the air generally also lead to the formation of other sulfur oxides (SO_x) that can react with other compounds in the atmosphere to form small particles thus, contributing to particulate matter pollution. Contradictly, Shah Alam and Nilai that are also industrial-based areas recorded very weak association of PM₁₀ with SO₂, with the r-value of -0.1 and 0.06. This might be due to the background of the station that is an urban-industry and commercial area where high correlation of NO_x-NO₂ were observed.

Other than that, weak association of meteorology parameters and other gaseous with PM_{10} level were observed. However, strong positive correlation between meteorology parameters such as wind speed and temperature were observed during HPE except in Larkin. Interestingly, significant correlation of O₃ level with the meteorology parameters were observed during HPE. Strong positive correlation was detected between O₃ level with wind speed and temperature whereas, strong negative correlation was detected with humidity. Ozone, a secondary pollutant, undergo photochemical reaction to exist in troposphere and it will



Figure 5. Correlation coefficient matrix of PM₁₀ level and other parameters

reached its peak during the highest solar radiation during the day. Due to this, O₃ has a strong positive correlation with temperature. Moreover, relative humidity affects ozone production via formation of hydroxyl. Thus, the concentration of ozone was higher when the relative humidity was lower because more hydroxyl radicals were converted into ozone (Pallavi and Chirashree, 2011). Higher O₃ concentrations together with other pollutants such as PM and nitrogen oxides during HPE would create a more harmful mixture (O₃-NO₂-PM_{2.5}) in ambient environments (Liu and Peng, 2017).

4. Conclusion

The pattern and behavior of PM_{10} concentration at four urban-industrial areas located at the west coast of the peninsular Malaysia during episodic HPE (1997, 2004, 2013 and 2015) were analyzed. Generally, the mean value during all respective year were observed higher than the median value indicating the probability of extreme event. A significant peak of PM_{10} level indicating HPE for the observed year generally started in June or during the southwest monsoon until the inter-monsoon in October. Seasonal mapping distribution also show that high monthly average concentration was observed during the end of southwest monsoon

especially in August and September. The most affected areas were Bukit Rambai, Nilai and Shah Alam whereas Larkin seems to have moderate to less effect of HPE. Trajectory analysis also show that HPE during 2005 and 2013 was originated by the forest fire events occurs in Sumatra region while 2015 from Kalimantan region. Generally, strong positive correlation was observed between PM₁₀ and CO concentrations whereas very weak relationship of PM₁₀ level with meteorological parameters were detected. However, high correlation between O₃ level and meteorology parameters were detected during HPE. Noticeable higher concentration of O₃ level during HPE was reported due to many factors including photochemical reactions of O₃ via formation of hydroxyl ion and the light scattering due to particles during HPE. Further studies on relationship of particles with O₃ level and its precursors, NO₂ during HPE is urgently needed as the O₃-NO₂-PM_{2.5} was reported to be more harmful to human health compared to particulate matter alone. These findings is significant in providing better understanding on the episodic haze episodes in Malaysia. It is anticipated that by revealing comprehensive analysis of PM₁₀ level during HPE will be an assistance to the authorities for policy/strategies development in Malaysia, in hope to reduce haze severity during future episodes.

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