



## Variability of PM<sub>10</sub> level with gaseous pollutants and meteorological parameters during episodic haze event in Malaysia: Domestic or solely transboundary factor?

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### ABSTRACT

Haze has become a seasonal phenomenon affecting Southeast Asia, including Malaysia, and has occurred almost every year within the last few decades. Air pollutants, specifically particulate matter, have drawn a lot of attention due to their adverse impact on human health. In this study, the spatial and temporal variability of the PM<sub>10</sub> concentration at Kelang, Melaka, Pasir Gudang, and Petaling Jaya during historic haze events were analysed. An hourly dataset consisting of PM<sub>10</sub>, gaseous pollutants and weather parameters were obtained from Department of Environment Malaysia. The mean PM<sub>10</sub> concentrations exceeded the stipulated Recommended Malaysia Ambient Air Quality Guideline for the yearly average of 150 µg/m<sup>3</sup> except for Pasir Gudang in 1997 and 2005, and Petaling Jaya in 2013. The PM<sub>10</sub> concentrations exhibit greater variability in the southwest monsoon and inter-monsoon periods at the studied year. The air masses are found to be originating from the region of Sumatra during the haze episodes. Strong to moderate correlation of PM<sub>10</sub> concentrations was found between CO during the years that recorded episodic haze, meanwhile, the relationship of PM<sub>10</sub> level with SO<sub>2</sub> was found to be significant in 2013 with significant negatively correlated relative humidity. Weak correlation of PM<sub>10</sub>-NOx was measured in all study areas probably due to less contribution of domestic anthropogenic sources towards haze events in Malaysia.

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## 1. Introduction

Haze is a mixture of pollutants transported by air across the border, that includes soot particles, carbon dioxide, and other toxic gases, where the horizontal visibility of less than 10 km is deteriorated [1–3]. Haze is influenced by meteorological conditions and physical or chemical interactions [4,5]. It is associated with industrial activities, traffic emissions, and biomass burning [6,7]. The air quality in Malaysia has been degraded due to the annual open biomass burning in Sumatra, Indonesia which resulted in a trans-boundary haze. Forest fires in Indonesia have intensified due to El Niño, improper forest management, and fire control [1]. An El Niño takes place when the temperature of the surface water in the equatorial Pacific becomes warmer than average and the east winds blowing weaker than normal [8], often leads to prolonged and drier weather conditions in the region [9]. The expansion of trans-boundary air pollution and historic haze events has become a huge concern in Malaysia [10–12]. Malaysia lies within the main pathway of the Southeast Asian pollution outflow which has allowed significant exposure to regional aerosol and pollutant emissions [13,14]. The southern and central regions of the Malaysian Peninsula have been severely affected by the transboundary haze originating from Sumatra, Indonesia, where high concentrations of particulate matter were emitted from the open burning [13,15]. Several episodes of severe haze have been reported whereby the concentrations of PM<sub>10</sub> exceeded the Recommended Malaysian Ambient Air Quality Guideline (RMAAQG) at various locations. The history of haze episodes in Malaysia were recorded in the year 1997, 2005, 2013, and 2015. The impact of vegetation fires on ambient air quality in Southeast Asia had become noticeable by July 1997 and it rose in September and started to decline when the rainy season began in November [16–18]. The significant haze outbreak reported in 2005 occurred primarily on peninsular Malaysia's central west coast [12,19]. It occurred regularly almost every year during the dry season since its occurrence in 2005. Klang Valley and its surrounding areas were badly affected by the smoke haze. The haze in 2013 has severely affected the majority of peninsular Malaysia. Department of Environment [20] reported that the hazardous level of API reading was recorded in Muar District, Johor. The September 2015 haze episode was the longest ever episode recorded in Malaysia [21]. Exposure to haze has been associated with respiratory-related diseases, with particulate matter being a prominent air pollutant released during haze episodes. Hospital admissions due to respiratory health issue have been recorded during haze episodes [22]. An eight percent increase observed during the 1997 haze episode in Kuching compared to the average admissions in the same month of 1995, 1996, and 1998 [23].

Various studies have been conducted to study the spatial-temporal variation of PM<sub>10</sub> concentration across Malaysia. Mohamad Khir et al. [24] conducted a study on the spatial-temporal variation to identify the trend of PM<sub>10</sub> concentrations in the Southern Peninsular of Malaysia from year 2005–2015. The finding of the study indicates that only year 2011 and 2012 did not experience transboundary haze. The highest PM<sub>10</sub> concentration was observed during the dry season in March, while the lowest concentration was observed during the monsoon season in December. A research by Tella et al. [25] utilised the Geographic Information System (GIS), a multi-variate regression model (MVR), and Pearson correlation analysis to study the relationship between PM<sub>10</sub> and meteorological parameters in Malaysia. The study found that temperature, wind speed, and humidity were identified as the most crucial factors influencing the concentration of PM<sub>10</sub> in the area of interest. Asmat et al. [26] and assessed the spatio-temporal variation of PM<sub>10</sub> in Klang Valley by using Inverse Distance Weighted (IDW) technique at different monsoon seasons and meteorological conditions. The study found that the variation of monsoon seasons resulted in the variation in meteorological conditions which affected the distribution of PM<sub>10</sub> especially during the southwest monsoon season. A study of spatio-temporal variation of PM<sub>10</sub> across Malaysia conducted by Juneng et al. [27] concluded that PM<sub>10</sub> variability in Malaysia has four dominant modes in which each describing different spatial and temporal variations. Though, the first mode (the most dominant reported that PM<sub>10</sub> showing a peak concentration during the summer monsoon (southwest monsoon) and a minimal concentration during the winter monsoon (northeast monsoon), which is consistent with the previous findings [24,26].

All of these studies on PM<sub>10</sub> variability with gaseous pollutants and meteorological parameters were conducted during the normal condition of atmospheric pollutants or during non-haze events. Hence, the lack of information, especially on the relationship of PM<sub>10</sub> level during the high particulate event (HPE) with the gaseous and weather parameters. Understanding on the distribution and correlation of PM<sub>10</sub> concentration with the gaseous and weather parameters during HPE are crucial as it provides scientific information that are reliable for policy makers to make decision. In addition, the influence of weather patterns, such as wind speed and direction, was not taken into consideration in most of the studies on the spatio-temporal distribution of PM<sub>10</sub> especially during haze event. Therefore, the findings may not accurately reflect the impact of weather patterns on PM<sub>10</sub> concentrations in the region.

Therefore, the main focus of our study is to evaluate and compare the characteristics and behaviour of PM<sub>10</sub> levels specifically during the episodic haze event (in 1997, 2004 2013 and 2015). Besides, it is aim to investigate whether domestic anthropogenic factor has any influence during the episodic haze event in Malaysia. Though, the conclusion for this investigation was made only using statistical approach due to very limited information were found on chemical apportionment of the smoke/PM during the haze event in Malaysia. The results from this study may be useful as it provides a new insight information during episodic haze events, thus the local

**Table 1**  
The study locations and its background.

Monitoring station	Representative Location	Latitude (N)	Longitude (E)	Background	Population [28]
Sekolah Menengah (P) Raja Zarina	Kelang	3.01217°	101.40978°	Urban	1,088,942
Sekolah Menengah Tinggi Melaka	Melaka	2.19210°	102.25449°	Urban	597,135
Sekolah Menengah Pasir Gudang 2	Pasir Gudang	1.47106°	103.89546°	Industrial	1,711,191
Sekolah Kebangsaan Bandar Utama	Petaling Jaya	3.13217°	101.60814°	Industrial	2,298,130

authorities in Malaysia are aware on the latest information.

## 2. Materials and methods

### 2.1. Study area

The locations selected are located on the west coast of peninsula Malaysia. The specific locations of the study are Kelang, Melaka, Pasir Gudang, and Petaling Jaya. All of these stations were prone to transboundary smoke from the Sumatra region as these stations reside on the west coast of peninsular Malaysia. The locations of the study and its background are shown in Table 1 and Fig. 1.

### 2.2. Air pollutant data

Datasets from the Department of Environment (DOE) Malaysia at the four locations mentioned above were obtained from the year when severe haze is reported, which were in 1997, 2005, 2013, and 2015. 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. The procedures that were used in the monitoring stations followed the standards outline States Environmental Protection Agency (USEPA) [29]. The data that were acquired from the DoE, Malaysia was usually comply with standard quality control processes and quality assurance procedures [30]. The procedures that were used in the monitoring stations followed the standards outline States Environmental Protection Agency (USEPA) [31].

The dataset consists of hourly data on PM<sub>10</sub> concentrations, gaseous pollutants such as nitrogen oxides (NO<sub>x</sub>), sulfur dioxide (SO<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>), ozone (O<sub>3</sub>), and carbon monoxide (CO) as well as meteorological parameters such as wind speed, relative temperature, and humidity. Table 2 shows the air pollutants and weather parameters that were obtained from DOE, Malaysia.

### 2.3. Spatio-temporal variation of PM<sub>10</sub> concentration

Dataset of the PM<sub>10</sub> concentrations, gaseous pollutants, and weather parameters are analysed via descriptive analysis. The descriptive analysis was conducted using IBM SPSS Statistic 26. The measure of central tendency, which includes mean, median, and mode, is used to study the central value in the distribution of the dataset obtained. Additionally, the variability of the data is interpreted using the measure of dispersion, which includes standard deviation and range. Time-series plot of PM<sub>10</sub> concentration illustrated the trend of PM<sub>10</sub> concentrations annually for the years 1997, 2005, 2013, and 2015 at all study locations. Box plot summarised the information about the shape, dispersion, and centre of the data.

A contour plot is constructed to indicate the spatial variation of the PM<sub>10</sub> during the high particulate event (HPE). The contour plot is developed by using the ArcMap 10.4 software. The data required to develop the contour plot are the location coordinates (x and y) as well as the value of the PM<sub>10</sub> concentrations (z) at the respective locations. Kriging interpolation method is chosen from the Spatial

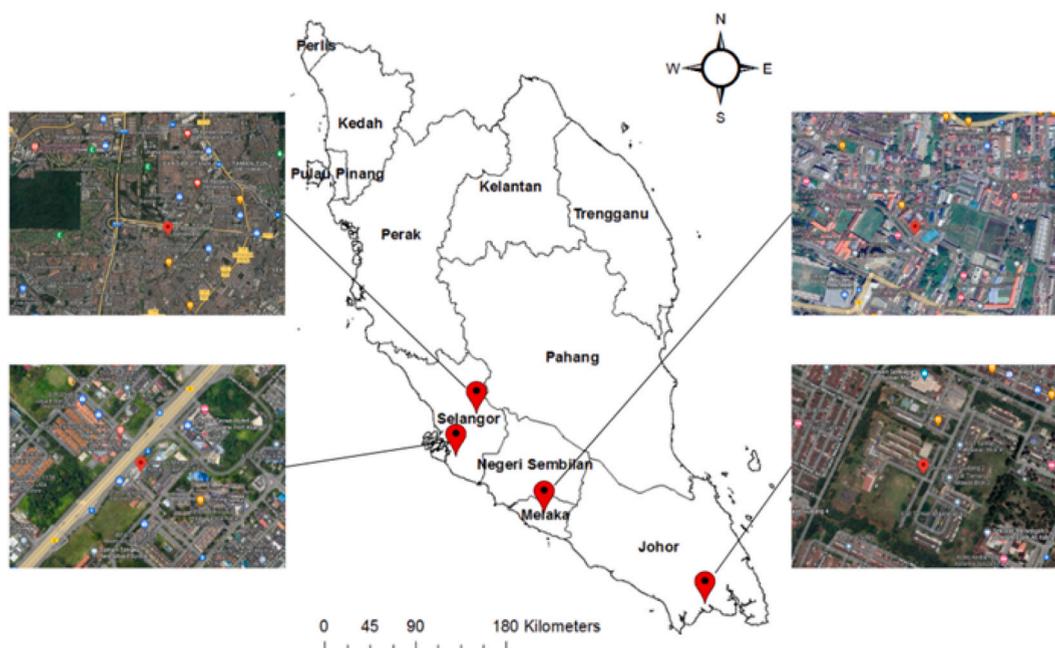


Fig. 1. The locations of the study.

**Table 2**  
List of air pollutants and weather parameters.

Parameters		Abbreviation	Unit
Air pollutants	Particulate matter	PM <sub>10</sub>	µg/m <sup>3</sup>
	Nitrogen oxides	NO <sub>x</sub>	ppm
	Sulfur dioxides	SO <sub>2</sub>	ppm
	Nitrogen dioxides	NO <sub>2</sub>	ppm
	Surface ozone	O <sub>3</sub>	ppm
Weather parameters	Temperature	T	°C
	Wind speed	WS	km/h
	Relative humidity	H	%

Analyst's Tools to predict and assess the distribution of PM<sub>10</sub> to other regions. This interpolation technique uses the known locations coordinate to determine the value of PM<sub>10</sub> at the unknown area [25]. The characterised mean and the variance provides the basis for the prediction of the unknown area [32].

A trajectory modeling is computed via The National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory's (ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) to assess the origin of air masses transported to the selected study locations during historic haze events. Is used in the modeling. The combination of the Lagrangian and Eulerian techniques has produced the model by adopting a moving frame of reference for the advection and diffusion calculations as the trajectories of an air mass from a specific location and at the same time computing the air pollutant concentrations using a fixed three-dimensional grid as a frame of reference [33]. The archive backward trajectories are computed by specifying the dates where the worst haze were recorded for each year and by using the 120-h (four days) at the height of 500 m above ground level (AGL). The archived datasets used in the computation is from GDAS (1°, global, 2006-present). The results are downloadable once the system generated the trajectories.

#### 2.4. Pearson's correlation analysis

The association between PM<sub>10</sub> concentration with gaseous pollutants and weather parameters at Kelang, Melaka, Pasir Gudang, and Petaling Jaya are identified using Pearson's correlation analysis, which is also known as Pearson's correlation,  $r$ . The PM<sub>10</sub> concentration used in this analysis was limited to concentrations that were  $>100 \mu\text{g}/\text{m}^3$  as stipulated in New Malaysia Ambient Air Quality Standard thus the correlation evaluated the relationship of high concentration of PM<sub>10</sub> with other parameters. The correlation analysis for each location is conducted by using IBM SPSS Statistic 26. The coefficient,  $r$  value ranges between +1 and -1. A positive correlation is shown by a value larger than 0, whereas a negative association is indicated by a value less than 0. Table 3 indicated the interpretation of the size of a correlation coefficient.

The value of  $r$  can be calculate using the given formula [35]:

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

where  $x_1, x_2, x_3, \dots, x_n$  and  $y_1, y_2, y_3, \dots, y_n$  represent the individual data value of  $x$  and  $y$  variables, respectively, and  $n$  is the observation number.

### 3. Results and discussion

#### 3.1. Descriptive statistics

The data summary for PM<sub>10</sub> concentrations at each study area in 1997, 2005, 2013, and 2015 is summarised in Table 4.

Overall, Kelang recorded the highest mean in all four years. The transboundary haze originating from the land fires in Sumatra, Indonesia contributed to the high mean value of PM<sub>10</sub> concentrations in Kelang. The burning vegetation contains high levels of ambient particulate matter. A study by Engling et al. suggested that the total suspended particles level increased during the haze episode, caused by biomass burning and peat fires in Indonesia compared to normal days [36]. Engling et al. used a chemical mass balance approach and found that the total suspended particles were mainly contributed by the peat fires followed by diesel exhaust and ship emissions during the peat fires in Indonesia [36]. Additionally, Melaka showed the highest mean as well in 2015. The mean in

**Table 3**  
Interpretation of Pearson's correlation coefficient value,  $r$  [34].

Size of correlation ( $r$ )	Interpretation
0.6 to 1.0	High correlation
0.3 to 0.6	Medium correlation
0.0 to 0.3	Low correlation

**Table 4**  
Data summary of PM<sub>10</sub> concentrations.

		Kelang				Melaka			
		1997	2005	2013	2015	1997	2005	2013	2015
N	Valid	8631	8744	8653	8622		7427	8163	8622
	Missing	129	16	107	138		1333	597	138
Mean		87.41	78.54	64.79	76.96		57.13	57.24	76.96
Std. Deviation		59.90	53.09	43.00	48.18		20.05	43.75	48.18
Range		396.00	619.00	573.00	388.00		160.00	497.00	388.00
Minimum		18.00	24.00	22.00	0.00		18.00	18.00	0.00
Maximum		414.00	643.00	595.00	388.00		178.00	515.00	388.00
Percentiles	25	46.00	54.00	47.00	52.00		45.00	40.00	52.00
	50	70.00	68.00	58.00	64.00		52.00	50.00	64.00
	75	102.00	86.00	70.00	82.00		64.00	60.00	82.00
	80	114.00	92.00	74.00	90.00		66.00	64.00	90.00
	90	162.00	116.00	88.00	120.00		78.00	76.00	120.00
		Pasir Gudang				Petaling Jaya			
		1997	2005	2013	2015	1997	2005	2013	2015
N	Valid	8631	8715	8745	8710	8222	8727	8659	8591
	Missing	129	45	15	50	538	33	101	169
Mean		47.72	46.59	50.99	64.80	69.42	64.32	48.43	58.76
Std. Deviation		39.87	13.70	38.43	36.09	55.05	40.70	29.31	50.48
Range		257.00	97.00	452.00	324.00	373.00	474.00	355.00	526.00
Minimum		11.00	19.00	10.00	27.00	20.00	20.00	17.00	5.00
Maximum		268.00	116.00	462.00	351.00	393.00	494.00	372.00	531.00
Percentiles	25	23.00	38.00	36.00	44.00	39.00	48.00	35.00	31.00
	50	33.00	44.00	45.00	54.00	49.00	56.00	43.00	43.00
	75	52.00	54.00	56.00	70.00	76.00	68.00	54.00	64.00
	80	62.00	58.00	58.00	78.00	86.00	72.00	58.00	74.00
	90	100.00	66.00	68.00	108.00	128.00	86.00	68.00	115.00

each year at each location is higher than the median signifying that the distribution of the data is positively skewed and there was an extreme event that occurred [37]. From the table, the 90th percentiles at each study location indicated at 90th percentile of PM<sub>10</sub> concentrations data did not exceed the Recommended Malaysia Ambient Air Quality Guideline (RMAAQG) for the hourly average of 150 µg/m<sup>3</sup> at each study location for each year except for Kelang in the year 1997. Thus, this indicates that the high particulate event might occurred due to high dispersion or effective wet scavenging. Wet scavenging is one of the most efficient processes for removing aerosols from the atmosphere [38]. This can be proved by the maximum concentration of PM<sub>10</sub> recorded in all areas during these years that exceeded 150 µg/m<sup>3</sup> except for Pasir Gudang in 2005.

Table 5 shows the tabulated values of the total number of unhealthy hours and days recorded in Kelang, Melaka, Pasir Gudang, and Petaling Jaya during the episodic haze event. The unhealthy hours and days were calculated based on the number of exceedances of PM<sub>10</sub> concentration that exceeded 100 µg/m<sup>3</sup>. Kelang recorded the highest number of unhealthy hours in 1997 where the exceedances occurred due to the transboundary haze pollution transported from the land burning in the Sumatra region of Indonesia which overlapped with the dry season [39]. A study by Mott et al. [23] suggested that the forest fire smoke in 1997 experienced short-term has led to an increase in cardiorespiratory hospitalisations. The number of daily outpatient visits for respiratory diseases in Kuala Lumpur General Hospital increased from 250 to 800 [40]. Awang et al. [41] reported that asthma cases in Selangor rose from only 912 in June to more than 5000 in September 1997. The total number of acute respiratory infection cases increased from about 6000 to more than 30000 during the same period. Melaka recorded the highest number of unhealthy hours in 2013 and 2015 due to massive land and forest fires in Sumatra and Kalimantan, Indonesia [42]. Haze event in 2015 recorded high number of hours and days that PM<sub>10</sub> exceeded 100 µg/m<sup>3</sup>. The highest hours was observed in Klang whereas the highest day was observed in Petaling Jaya. It can be seen that trend of unhealthy hours and days were similar to the 1997. Latif et al. [10] reported that haze event in 2015 were the longest

**Table 5**  
Hourly and daily PM<sub>10</sub> concentrations exceeding the RMAAQG (>100 µg/m<sup>3</sup>).

Station	1997		2005		2013		2015	
	TH	TD	TH	TD	TH	TD	TH	TD
Kelang	2388	144	1429	84	589	40	1406	77
Melaka	–	–	403	26	368	21	1103	96
Pasir Gudang	869	45	21	2	252	13	998	51
Petaling Jaya	1246	74	520	36	268	18	923	113

Note:

Total hours (TH) - number of hours that the PM10 concentration exceeds 100 µg/m<sup>3</sup>.

Total days (TD) - number of days with at least 1-h the PM10 concentration exceeds 100 µg/m<sup>3</sup>.

recorded event that lasted for about 2 months and it was stated as the worst haze period in Malaysia’s history after 1997.

3.1.1.1. Daily PM<sub>10</sub> level variation

To observe clearly the exact trend of haze events during these years, Fig. 2 shows the time-series plot for daily PM<sub>10</sub> concentration in the year 1997, 2005, 2013, and 2015 at Kelang, Melaka, Pasir Gudang, and Petaling Jaya. The Recommended Malaysia Ambient Air Quality Guideline (RMAAQG) for 24-h averaging time which is 100 µg/m<sup>3</sup> is represented by the red solid line. The highest concentrations observed in the year 1997 are at Kelang and Petaling Jaya on 15th September. The high concentrations of PM<sub>10</sub> at Kelang and Petaling Jaya continued from early September to the mid of September. The vegetation fires in Kalimantan and Sumatra resulted in high concentrations of PM<sub>10</sub> in Kelang and Petaling Jaya during September 1997, which occurred during May and June of the same year [43]. It coincided with the El Niño phenomenon, which prolonged the dry season in that year. A smoke-haze layer has formed in Malaysia due to transboundary pollution [16].

The bimodal peak of PM<sub>10</sub> concentrations are observed at Kelang and Petaling Jaya in 2005 on the 17th and 25th of February. Another peak concentration of PM<sub>10</sub> rose to start on 11th August. Sulaiman et al. [44] reported that the haze episode in August 2005 was more severe compared to the 1997 haze occurrence in peninsular Malaysia. The haze episode largely affected the entire Klang Valley and its nearby areas. The dense population and heavy traffic in the Klang Valley result in significant vehicle emissions, such as emissions of exhaust gases and congested traffic could possibly worsen air quality more in the area. It was observed that Melaka and Pasir Gudang were not affected by the haze event in 2005. The Air Pollution Index (API) at Kelang exceeded 500 thus a Haze

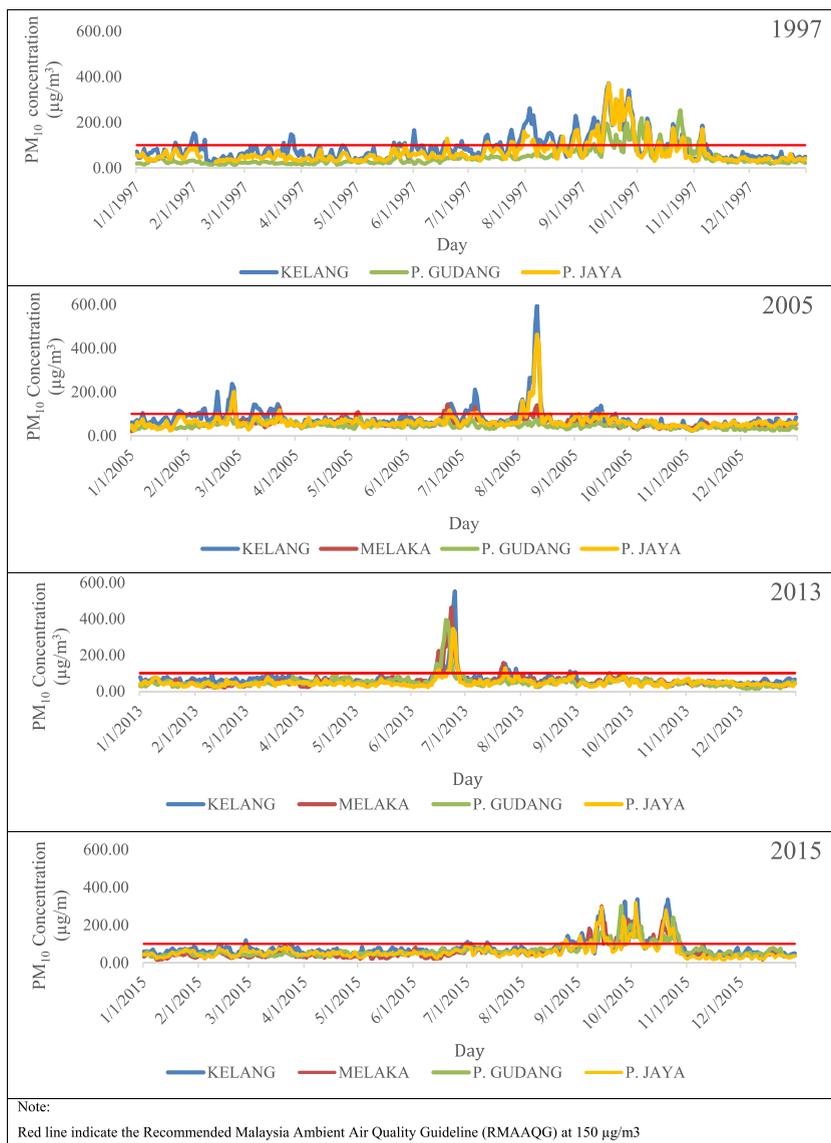


Fig. 2. Daily time-series plot for Kelang, Melaka, Pasir Gudang and Petaling Jaya.

Emergency was declared in the area [44]. High concentration of PM<sub>10</sub> in Petaling Jaya is observed because it is located in Klang Valley, which is the most developed area in Malaysia and has the largest population [44]. Amir [45] reported that vehicle exhaust is the primary source of air pollution in Petaling Jaya since it is a mixed commercial-residential-industrial area that has the highest population in the Klang Valley. The air quality in the area deteriorated not only due to transboundary haze, but it was also contributed by anthropogenic sources [45]. However, from the data summarised in Tables 3 and it was recorded that the concentration of PM<sub>10</sub> at the 90th percentile in Kelang and Petaling Jaya in 2005 did not exceed the value stipulated by RMAAQG (150 µg/m<sup>3</sup>), hence it can be concluded that transboundary air pollution was the only reason for extreme high particulate event in Klang Valley that year.

In 2013, the highest PM<sub>10</sub> concentrations observed at Kelang, Melaka, Pasir Gudang, and Petaling Jaya are on 25th June 2013, 23rd June 2013, 21st June 2013, and 24<sup>th</sup> June 2013, respectively. Malaysia experienced a short period of severe haze events episode from 15 to 27 June 2013 [46]. The air quality in most regions within peninsular Malaysia worsened as a result of the transboundary pollution transported from massive land burning in Sumatra, Indonesia [13]. All four locations were affected during the short period of the haze episode.

In 2015, it can be observed that the peak PM<sub>10</sub> concentrations at all four study locations started to rise from early September until the end of October 2015. PM<sub>10</sub> concentrations exceeded the RMAAQG with fluctuating trend between September and October in that year. The air quality in Malaysia deteriorated from August to October 2015 due to huge land and forest fires in Sumatra and Kalimantan, Indonesia [47]. It occurred during the period of southwest Monsoon. El Niño in 2015 was strong and a prolonged drought was observed in Southeast Asia [48]. The El Niño and drought resulted in the widespread of seasonal fires in Indonesia was greatly inflated which causes large amounts of terrestrially-stored carbon into the atmosphere [33]. Department of Environment [49] reported that for the first time in Malaysia's history since 1997, 34 locations in the country experienced unhealthy air quality levels on September 15, 2015.

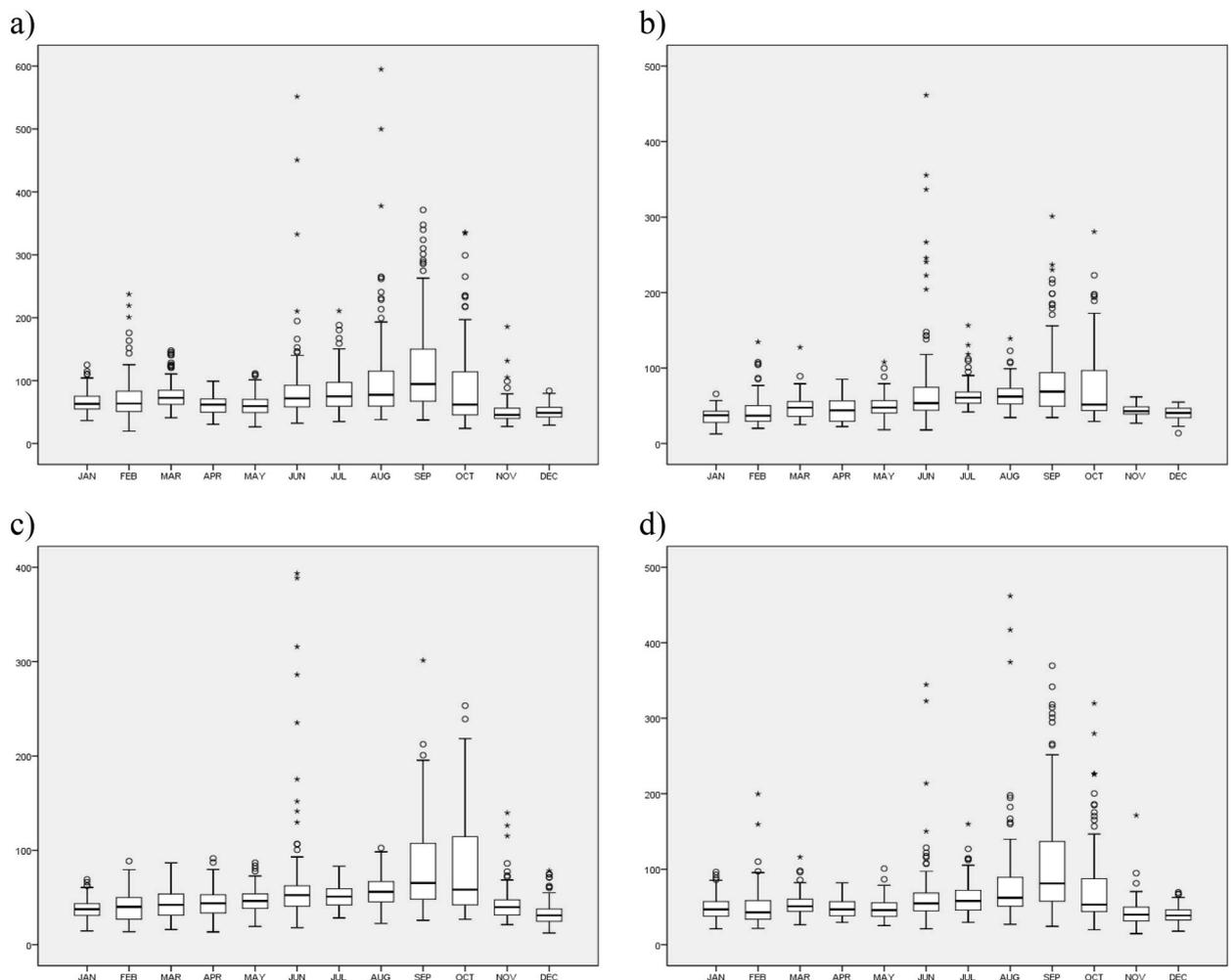


Fig. 3. Box plot distribution for PM<sub>10</sub> concentrations at a) Kelang, b) Melaka, c) Pasir Gudang and d) Petaling Jaya.

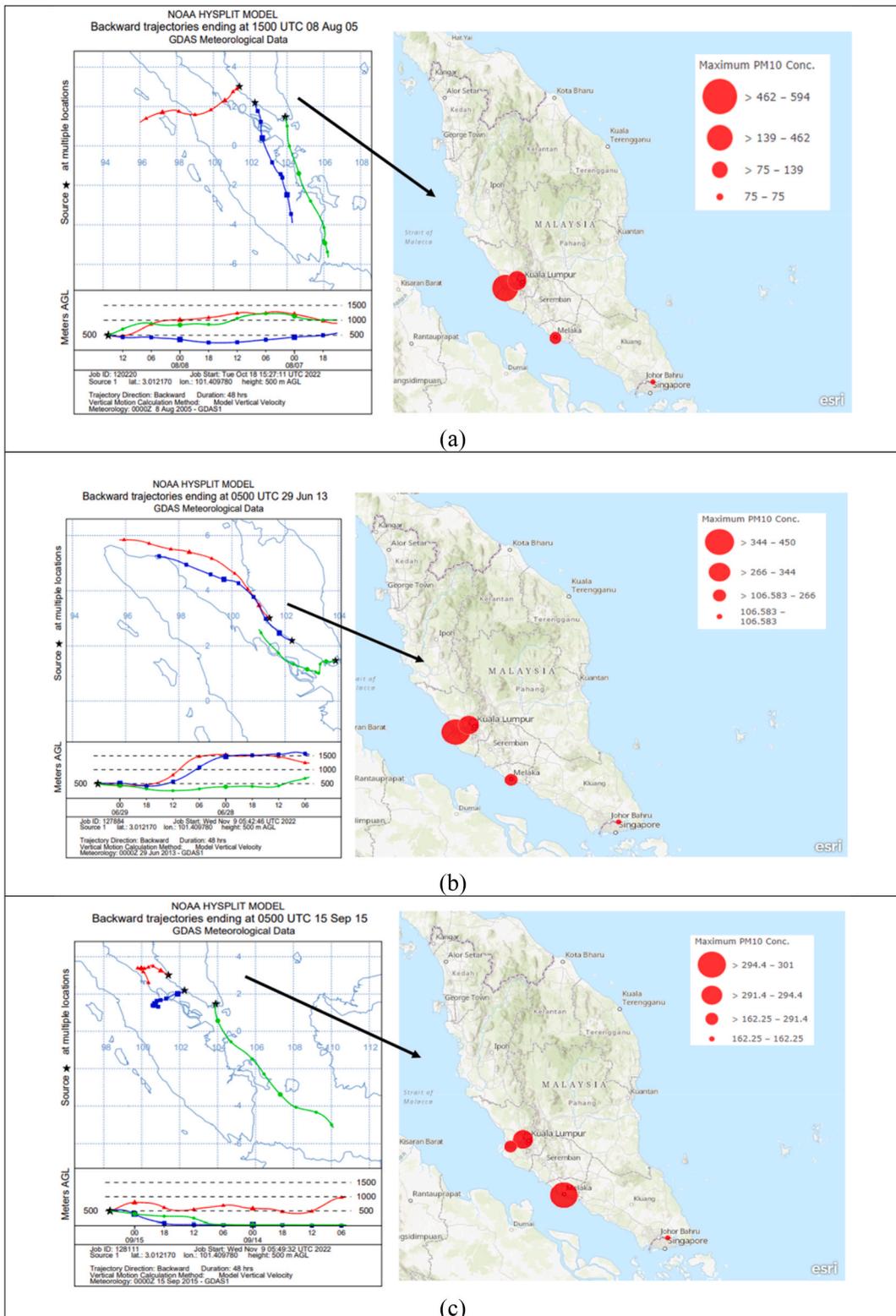


Fig. 4. The backward trajectories of wind and the spatial distribution of PM<sub>10</sub> concentration during haze event in (a) 2005; (b) 2013 and; (c) 2015.

### 3.1.2. Monthly PM<sub>10</sub> level variation

To observe the monthly distribution of PM<sub>10</sub> levels during the episodic haze event, box plots of PM<sub>10</sub> concentrations for all four locations were developed as shown in Fig. 3(a–d). Generally, the extreme PM<sub>10</sub> concentration can be observed during the southwest monsoon i.e. (from June to September) and inter-monsoon (October). Higher variability of PM<sub>10</sub> concentrations is recorded during September at Kelang and Petaling Jaya. Meanwhile, Melaka and Pasir Gudang showed very variable PM<sub>10</sub> concentrations in October. The wind presence during the southwest monsoon and biomass burning affects the concentration of air particulate matter in Southeast Asia, specifically Malaysia [50]. The biomass burning-related transboundary pollution originated from Indonesia. Studies by Juneng et al. [27] found that the maximum PM<sub>10</sub> concentration overlapped when regional low-level winds were primarily southerlies and south westerlies, as well as when the region experienced a dry season. The southwest monsoon's high PM<sub>10</sub> concentrations may be caused by the lack of precipitation and high temperatures [15].

### 3.1.3. Backward trajectory

To further characterize the spatial behaviour of PM<sub>10</sub> concentrations, Fig. 4 shows the backward trajectories of wind and the spatial distribution of PM<sub>10</sub> concentration during the haze event. The trajectories are calculated for 48 h at the height of 500 m above ground level. From Fig. 4(a), the trajectory plot indicated that the air masses traveled from the North Sumatra region to Kelang meanwhile, the air masses arriving at Melaka and Pasir Gudang originated from the South Sumatra region during the haze episode in August 2005. It was observed that Kelang was the most affected area with extremely high concentration of which was >500 µg/m<sup>3</sup>. HPE occurred during 2005 which is mainly on the central west coast of the Peninsular Malaysia [12]. Severe smoke haze episodes occurred in 11 August in two areas namely Pelabuhan Klang and Kuala Selangor with PM10 concentration exceeded 500 µg/m<sup>3</sup> and then shifted to the northern states by 13 August 2005 [46]. The air masses during the haze event in June 2013 are transported from the region of north Sumatra to all study areas, as demonstrated in Fig. 4(b). Highest concentration of PM10 was also observed in Kelang compared to other areas with concentration ranging from 450 to 75 µg/m<sup>3</sup>. During the occurrence of haze in September 2015, the air masses arriving in Kelang and Melaka originated from the region of North Sumatra, while in Pasir Gudang, the air masses traveled from the South Sumatra region (Fig. 4(c)). It was detected that all study areas recorded high concentration of PM<sub>10</sub> ranging from 301 to 162 µg/m<sup>3</sup> with the highest concentration was recorded in Melaka. Haze event in 2015 was classified as the worst that had been recorded in Malaysia's history which was lasted for two months [10]. According to Department of Environment [49], all schools in the states of Putrajaya, Kuala Lumpur, Selangor, Negeri Sembilan and Melaka were closed due to the Air Pollution Index (API) reading reaching to the category of unhealthy and very unhealthy air. In conclusion, the equatorial Southeast Asia (SEA) region experiences a dry season which causes a rise in fire events during the summer or southwest monsoon [10]. Hence, the presence of southerly and south-westerly winds allows the cross-boundary transportation of pollutants from the burning areas in Sumatra to SEA region.

**Table 6**

Pearson's correlation of PM<sub>10</sub> concentration with other parameters (correlation is significant at 0.05 level).

Year	Location	WS	T	H	NO <sub>x</sub>	SO <sub>2</sub>	NO <sub>2</sub>	O <sub>3</sub>	CO
1997	Kelang	-0.08	-0.01		-0.06	0.28	-0.05	0.12	0.40
	Melaka								
	Pasir Gudang	-0.16	-0.09		0.23	0.08	0.36	0.02	0.53
	Petaling Jaya	-0.07	0.01		-0.03	0.14	-0.07		
2005	Kelang	-0.09	-0.02	0.07	0.07	-0.15	0.06	-0.11	0.65
	Melaka	0.02	0.00	0.04	-0.08	-0.14	-0.02	0.16	0.24
	Pasir Gudang	0.50	0.31	-0.31	-0.67	0.35		0.14	-0.52
	Petaling Jaya	-0.15	-0.04	0.11	0.00	-0.18	-0.05	-0.10	0.89
2013	Kelang	0.00	0.16	-0.16	-0.22	0.61	-0.35	0.16	0.78
	Melaka		0.22	-0.37	-0.16	0.41	-0.36	0.01	0.69
	Pasir Gudang	-0.05	-0.14	-0.17	-0.11	-0.69	-0.24	0.05	0.79
	Petaling Jaya	-0.08	0.20	-0.14	-0.10	0.10		0.25	0.89
2015	Kelang		-0.03	0.09	-0.06	0.11	-0.09	0.02	0.56
	Melaka	0.05	0.11	-0.01	-0.06	-0.06	0.05	0.10	0.79
	Pasir Gudang	-0.12	0.09	0.15	0.09	0.21	0.14	-0.04	0.63
	Petaling Jaya	-0.02	0.09	-0.06	-0.21	0.08	-0.16	0.13	0.37

Note:

WS – wind speed.

T – temperature.

H – humidity.

NO<sub>x</sub> – nitrogen oxides.

SO<sub>2</sub> – sulfur dioxide.

NO<sub>2</sub> – nitrogen dioxide.

O<sub>3</sub> – ozone.

CO – carbon monoxide.

### 3.2. Association of PM<sub>10</sub> level with other gases and weather parameters

#### 3.2.1. Correlation analysis

Pearson’s correlation analysis was conducted to understand whether there is an association between PM<sub>10</sub> concentration with other air pollutants (NO<sub>x</sub>, SO<sub>2</sub>, NO<sub>2</sub>, O<sub>3</sub>, and CO) and weather parameters such as wind speed, temperature, and humidity during the high particulate event. Table 6 classified the relationship of PM<sub>10</sub> level with the other parameters according to year with the occurrence of the historic haze event.

Overall, PM<sub>10</sub> level in each location were found to have a high correlation with CO, SO<sub>2</sub>, and NO<sub>x</sub>. None of the meteorological parameters exhibit a significant correlation with PM<sub>10</sub> concentrations. In 1997, a moderate correlation was found only between PM<sub>10</sub> concentrations with CO in Kelang ( $r = 0.40$ ) and Pasir Gudang ( $r = 0.53$ ). A high correlation was observed between PM<sub>10</sub> and CO concentrations in Kelang ( $r = 0.65$ ) and Petaling Jaya ( $r = 0.89$ ) in 2005. Moreover, PM<sub>10</sub> and CO concentrations in Melaka and Pasir Gudang also showed a high correlation in the year 2005, with  $r$  values 0.79 and 0.63, respectively. PM<sub>10</sub> concentration exhibited a high correlation with CO at all four locations in the year 2013. A high correlation between PM<sub>10</sub> concentrations with CO levels may indicate

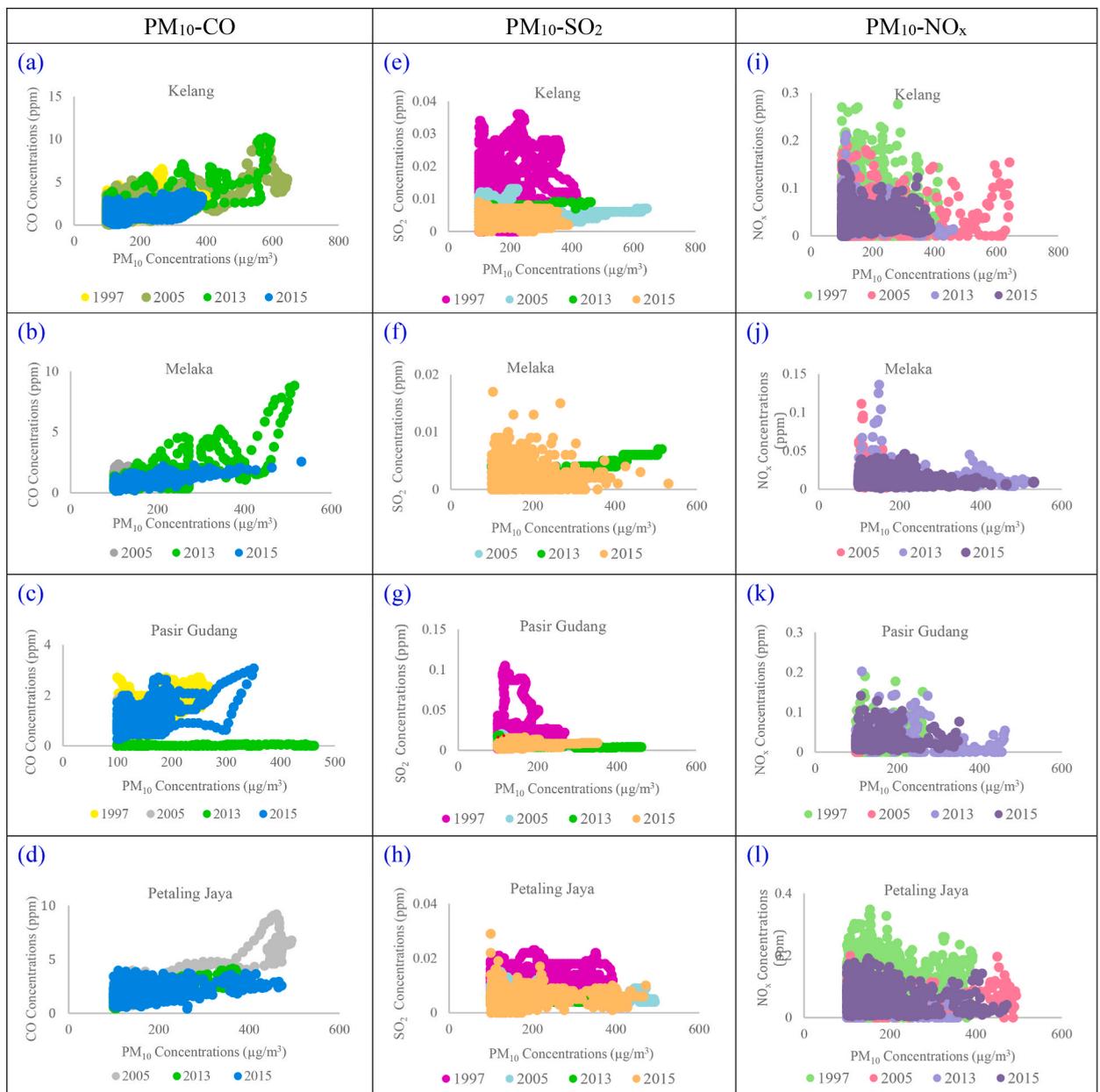


Fig. 5. Scatter plot of PM<sub>10</sub>-CO (a–d), PM<sub>10</sub>-SO<sub>2</sub> (e–h) and PM<sub>10</sub>-NO<sub>x</sub> (i–l) during the haze episodes.

the contribution from local anthropogenic sources such as emissions from traffic congestion and machinery usage due to the locations' urban and industrial background. Additionally, the seasonal land-burning activities in the Sumatra region of Indonesia may have led to the high correlation. The massive land fires released huge amounts of terrestrially-stored carbon into the atmosphere, primarily in the form of CO<sub>2</sub>, CO, and CH<sub>4</sub> [33]. CO could possibly be contributed by the incomplete combustion processes. Incomplete combustion occurs generally due to poor mixing of the air and fuel, insufficient residence time, insufficient temperature and low total excess air [51]. This can occur in vehicles, power plants, industrial processes, and residential heating systems when the fuel does not burn efficiently. During these events, smoke travels over large parts of Indonesia as well as Singapore and parts of Malaysia [52]. Much of the smoke is from peatland fires. Over half have been cleared and drained, in particular for plantation development (including oil palm and acacia for pulp and paper production). Drained, but still wet peat soils burn incompletely, at relatively low temperatures, which results in relatively high emissions of a mix of pollutants including particulate matter, carbon monoxide, and Polycyclic Aromatic Compounds (PACs) [52].

PM<sub>10</sub> was found to be highly correlated with SO<sub>2</sub> in Kelang ( $r = 0.61$ ) and Pasir Gudang ( $r = -0.69$ ), both in year 2013. High correlation between PM<sub>10</sub> concentration with SO<sub>2</sub> was found in year 2013 at Kelang ( $r = 0.608$ ) and Pasir Gudang ( $r = 0.694$ ). Meanwhile, a moderate correlation between PM<sub>10</sub> concentrations with SO<sub>2</sub> in the same year was found, with an  $r$ -value of 0.407. The influence of diesel-powered vehicles on particle levels is suggested by the high correlation coefficients between PM<sub>10</sub> and SO<sub>2</sub> [53]. Sulfate is a main component of ambient particulate matter (PM) in the urban environment during haze episodes [54]. Among the pollutants, sulfur dioxide (SO<sub>2</sub>) is an important precursor of sulfate and new atmospheric particle formation. In addition, high concentrations of SO<sub>2</sub> in the air generally also lead to the formation of other sulfur oxides (SO<sub>x</sub>) that can react with other compounds in the atmosphere to form small particles thus, contributing to particulate matter pollution [54]. Relative humidity (RH) above 80% can significantly promote SO<sub>2</sub> oxidation on the CaCO<sub>3</sub> particles and form the CaSO<sub>4</sub>·2H<sub>2</sub>O crystals. In contrast, at a relative humidity below 40%, only a few CaCO<sub>3</sub> particles can be converted to CaSO<sub>4</sub> particles.

A high correlation between PM<sub>10</sub> and NO<sub>x</sub> concentrations was found in Pasir Gudang in the year 2005, with an  $r$  value of  $-0.67$ . Pasir Gudang is known to be an area where shipbuilding, phytochemicals manufacturing, and other logistics and transportation are operated [54]. Apart from HPE episodes, the emission of PM<sub>10</sub> may be contributed to industrial activities in the area. NO<sub>x</sub> is emitted when fuel is burned at high temperatures, which may come from industrial activities in Pasir Gudang [55].

As for association of PM<sub>10</sub> concentration with weather parameters (temperature, humidity and wind speed), very small correlation values ( $r$ ) were calculated for all places. Temperature was observed to positively and negatively correlated with PM<sub>10</sub> concentration with the range of  $r < +0.25$  and  $r < -0.03$  respectively. Previous researchers found that good weather conditions when there is an increased temperature, are able to reduce the air pollutants level and improve air quality as particulate matter (PM) concentration significantly correlates to meteorological factors [1–4]. Though, a few studies especially located in China found that PM concentrations have a negative correlation with the wind speed, temperature, relative humidity, and precipitation [56,57]. The influence of temperature on the air pollutant concentration became more obvious as the latitude increased [2]. The correlation coefficient values also differ in different seasons, where the coefficient value in autumn/winter was significantly higher than in spring/summer [56,58]. PM<sub>10</sub> was found to have a low negative correlation with relative humidity in all other places except for Melaka (2013). Humidity causes particles to become moist, thereby increasing their weight and reducing their diffusion [2], hence it is usually negatively correlated with particle. As for wind speed, most of the study areas have weak negative relationship with PM<sub>10</sub> concentration except in Melaka and Pasir Gudang where both of the areas are located near by the sea. A negative correlation between PM and wind speed was because the particulate matter had a tendency to remain close to the ground when there is no presence of wind [2]. However, the presence of strong wind in summer caused the dust to be suspended, which resulted in the positive correlation between PM and wind speed [3].

### 3.2.2. Scatter plot

From the above explanation, it was detected that there were three main gaseous pollutants that were highly or moderately correlated with PM<sub>10</sub> levels. In order to observe the linear relationship between the gaseous pollutants and PM<sub>10</sub> level, scatter plots for

**Table 7**  
R<sup>2</sup> value of linear relationship of PM<sub>10</sub>-CO, PM<sub>10</sub>-SO<sub>2</sub> and PM<sub>10</sub>-NO<sub>x</sub>.

Location	Year	PM <sub>10</sub> -CO	PM <sub>10</sub> -SO <sub>2</sub>	PM <sub>10</sub> -NO <sub>x</sub>
Kelang	1997	0.156	0.0631	0.0053
	2005	0.4262	0.0232	0.0021
	2013	<b>0.6042</b>	<b>0.3699</b>	0.0483
	2015	0.3097	0.0128	0.0002
Melaka	2005	0.0594	0.0192	0.0056
	2013	0.477	0.01658	0.0241
	2015	<b>0.625</b>	0.0039	0.0036
Pasir Gudang	1997	0.2823	0.0065	0.046
	2005	0.2685	0.1206	<b>0.1129</b>
	2013	0.0016	<b>0.4818</b>	0.0796
	2015	0.3906	0.0441	0.0235
Petaling Jaya	1997		0.0195	0.0001
	2005	<b>0.7867</b>	0.033	0.0003
	2013	<b>0.7889</b>	0.010	0.0513
	2015	0.1396	0.0058	0.0436

each correlation of  $PM_{10}$ -CO,  $PM_{10}$ -SO<sub>2</sub>, and  $PM_{10}$ -NO<sub>x</sub> were plotted and depicted in Fig. 5(a–l), where the R<sup>2</sup> for the correlation is given in Table 7. For  $PM_{10}$ -CO correlation (Fig. 5(a–d)), it can be observed that the correlation was observed highest in Petaling Jaya and Kelang during the 2013 haze event with R<sup>2</sup> values of 0.79 and 0.60 respectively. Petaling Jaya also recorded a high correlation of  $PM_{10}$ -CO during the 2005 haze event. For the haze event in 2015, the highest correlation of  $PM_{10}$ -CO was only observed in Melaka with the R<sup>2</sup> value of 0.63. Even though it was reported that the haze event in 1997 and 2015 had recorded a greater impact compared to other haze events, the correlation of  $PM_{10}$ -CO in these years was observed less great compared to 2013.

For the relationship of  $PM_{10}$ -SO<sub>2</sub> (Fig. 5(e–h)), it can be seen that the relationship was moderately correlated only during 2013 in Kelang and Pasir Gudang with the R<sup>2</sup> value of 0.48 and 0.37. During these haze episodes, sulfur dioxide (SO<sub>2</sub>) can be an important precursor of sulfate and new atmospheric particle formation. Higher humidity will promote higher SO<sub>2</sub> oxidation on the CaCO<sub>3</sub> particles and form the CaSO<sub>4</sub>·2H<sub>2</sub>O crystals [54]. From Table 6, the humidity was measured as one of the significant negatively correlated parameters that related to  $PM_{10}$  level in Kelang and Pasir Gudang with *r* values of −0.16 and −0.17. Thus, in this case, it might have a significant contribution to the correlation of  $PM_{10}$ -SO<sub>2</sub> in Kelang and Pasir Gudang during the 2013 haze.

Meanwhile, for the  $PM_{10}$ -NO<sub>x</sub> relationship (Fig. 5(i–l)), it only gives weak R<sup>2</sup> values of 0.13 which was measured at Pasir Gudang in 2005. Hence, it can be understood that the  $PM_{10}$ -NO<sub>x</sub> relationship was due to domestic contribution due to a very weak correlation during other haze events in all study areas. In the study areas, NO<sub>x</sub> and NO<sub>2</sub> were found out to have medium to strong correlation with  $PM_{10}$  in Kelang and Pasir Gudang (Table 6), however, it shows very weak fit of linear relationship with  $PM_{10}$  (R<sup>2</sup> > 0.2). NO<sub>x</sub> is a combination of NO and NO<sub>2</sub> and it is often used as tracer of road traffic emissions at monitoring sites located in urban areas [59]. NO<sub>x</sub> are also known as one of the main constituents involved in the formation of ground-level ozone, nitrate particles and acid aerosols which can trigger chronic respiratory diseases [60]. The relationship of NO<sub>x</sub>-ozone was not a linear relationship and current studies has proven that NO<sub>2</sub> that converted to nitrate (NO<sub>3</sub>) is an important component of PM<sub>2.5</sub> in major cities in China [48]. Therefore, further research need to be carried out to further understand the relationship of PM-NO<sub>x</sub> in the study area.

#### 4. Conclusion

The spatial-temporal trend of  $PM_{10}$  levels during historic haze events in Malaysia was evaluated, in years 1997, 2005, 2013, and 2015. Overall, the mean value of  $PM_{10}$  concentrations at all study areas exceeded the Recommended Malaysian Ambient Air Quality Guideline (RMAAQG) for the yearly average except for Pasir Gudang in 1997 and 2005 as well as Petaling Jaya in 2013. Kelang recorded the highest numbers of unhealthy hours where  $PM_{10}$  concentration exceeded the RMAAQG in years 1997 and 2005 resulting from the transboundary haze pollution transported from the event of fire in Indonesia which coincided with the dry season. Melaka recorded the highest numbers of unhealthy hours in 2013 and 2015 whereas the highest numbers of unhealthy days in 1997 and 2005 were observed in Kelang, and Melaka showed the highest numbers of unhealthy days in 2013 and 2015.  $PM_{10}$  concentrations varied during the period of southwest monsoon where the highest concentration was observed in September when Kelang consistently recorded the highest monthly average of  $PM_{10}$  level during the southwest monsoon (June to September) and inter-monsoon (October) period. The backward trajectory indicated that the air masses were transported from the northern and southern of Sumatra, Indonesia.  $PM_{10}$  concentrations in each study location were found to have a high and moderate correlation with CO for all haze events and a very weak correlation with weather parameters was detected. A high correlation between  $PM_{10}$  concentration with SO<sub>2</sub> was found to occur during the 2013 haze event, especially in Kelang and Pasir Gudang, meanwhile, the  $PM_{10}$ -NO<sub>x</sub> correlation was detected as weak during these episodic haze events. Further investigation is needed to confirm PM-NO<sub>x</sub> relationship as it has a nonlinear relationship. In this study, statistically it was proven that high particulate event in Malaysia was majorly related to the transboundary air pollutants from Indonesia and only a weak relationship between local anthropogenic sources was observed. However, to confirm the findings from this study, experimental analysis of chemical speciation of PM during the haze event need to be conducted. Due to very limited information on the chemical apportionment of PM during haze event in Malaysia, we could not validate these results with the actual situation in Malaysia.

Nevertheless, the findings of this study are essential in providing a better understanding of the influences of gaseous pollutants on the variation of  $PM_{10}$  concentrations during a historic haze episode in Malaysia. It will help the future locals and assist the authorities to enact preventative measures in order to reduce the impact of haze events. The results of this study provide a new insights regarding the relationship of  $PM_{10}$  concentration with other gaseous and weather parameters at major urban areas during historic haze event in Malaysia. Urban air pollution is an issue that must be addressed adequately to avoid health complications towards citizen since heavy industry impacts public health through the emission of many potentially hazardous pollutants into the air. One way to tackle this issue is by identifying the relationship between air quality and public health impacts. Hence, the information can be used by the epidemiological study to measure the impact of pollution to human wellbeing and the control strategies can be planned by the authorities.

#### Author contribution statement

Conceived and designed the experiments; Norazian Mohamed Noor; Ahmad Zia Ul-Saufie; Sandu Andrei Victor; Gyorgy Deak.

Performed the experiments; Nur Alis Addiena A Rahim; Izzati Amani Mohd Jafri.

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Contributed reagents, materials, analysis tools or data; Mohd Remy Rozainy Mohd Arif Zainol.

Wrote the paper; Nur Alis Addiena A Rahim; Norazian Mohamed Noor.

## Data availability statement

The data that has been used is confidential.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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