Spatiotemporal Variations of Particulate Matter and their Association with Criteria Pollutants and Meteorology in Malaysia

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Abstract

Fine particulate matter (PM\textsubscript{2.5}) poses a higher risk to human health than coarse particulate matter (PM\textsubscript{10}). This study aims to determine the spatiotemporal variations of PM\textsubscript{2.5} and PM\textsubscript{10} in Malaysia and their association with other criteria pollutants and meteorological factors. Hourly data from air quality monitoring stations for the year 2018 were retrieved from the Malaysian Department of Environment and analysed for temporal and spatial scales according to different regions in Malaysia. Further statistical analyses, such as Spearman’s Rank Correlation and Principal Component Analysis (PCA), were conducted to study the associations between PM\textsubscript{2.5} and PM\textsubscript{10} with other main criteria air pollutants, as well as meteorological parameters. Higher mean concentrations of PM\textsubscript{2.5} (23 ± 8 µg/m\textsuperscript{3}, range = 4.6 – 158 µg/m\textsuperscript{3}) and PM\textsubscript{10} (32 ± 10 µg/m\textsuperscript{3}, range = 6.0 – 181µg/m\textsuperscript{3}) were observed in the central region of the Malaysian Peninsula. The diurnal patterns of PM\textsubscript{2.5} and PM\textsubscript{10} were in a bimodal pattern and influenced by traffic emissions. The highest mean PM\textsubscript{2.5} and PM\textsubscript{10} concentrations were recorded during the southwest monsoon season, notably in the central region. The Spearman’s Rank Correlation shows that NO\textsubscript{2} and CO have a moderately positive correlation (\(p < 0.01\)) with PM\textsubscript{2.5} (\(r = 0.47\)) and PM\textsubscript{10} (\(r = 0.48\)) in the central regions while all meteorological parameters show significantly weak to very weak correlations with PM. The PCA analysis indicates that the major sources leading to the formation of particulate matter are from the contribution of secondary aerosols and combustion-related sources. The ratio of PM\textsubscript{2.5} to PM\textsubscript{10} ranged between 0.51 and 0.76 nationwide with the highest mean recorded in the central region (0.72). This study indicates that there is a higher abundance of fine particulate in the
ambient air of the urbanised environment and thus a greater likely risk to human health in more
developed areas.

**Keywords:** Particulate matter; Spatiotemporal; Atmospheric gases; Principal Component Analysis;
Ratio PM$_{2.5}$ to PM$_{10}$
Particulate matter is one of the major air pollutants in ambient air. Starting from the measurement of total suspended particulate (TSP), the measurement of particulate matter has progressed from particles with an aerodynamic diameter of less than 10 µm (PM\textsubscript{10}) to particles with an aerodynamic diameter lower than 2.5 µm (PM\textsubscript{2.5}) (Marcazzan et al., 2001). Most countries in the world have now started to use PM\textsubscript{2.5} as an additional air pollutant when determining the Air Pollution Index (Mukherjee and Agrawal, 2017; Wu et al., 2017). Due to its size, PM\textsubscript{2.5} can remain suspended in the atmosphere for a longer period of time and therefore has the capacity to travel a greater distance to downwind areas (Saunders and Waugh, 2015). Several studies have shown that PM\textsubscript{2.5} not only induces cardiopulmonary disorders and impairments but also contributes to a variety of adverse health effects, including the initiation and progression of diabetes mellitus and birth complications (Kappos et al., 2004; O'Neill et al., 2005; Brook et al., 2010; Feng et al., 2016; Zhang et al., 2020).

Fine particle PM\textsubscript{2.5} has different sources and compositions compared to coarse particle PM\textsubscript{10} (Marcazzan et al., 2002). Most coarse particles are directly emitted from sources such as crustal matter oxides (soil dust), fine particles, however, can be formed indirectly via the formation of nuclei through the vaporisation of heavy metals or the chemical reactions of gases (Lighty et al., 2000). These nuclei then undergo coagulation or condensation to form fine particulates. Coagulation occurs when particles combine to form larger particles while condensation occurs when gas or vapour molecules condense on the nuclei. The ratio between PM\textsubscript{2.5} and PM\textsubscript{10} can suggest the sources and the formation process of the PM emissions (Gehrig and Buchmann, 2003; Blanco-Becerra et al., 2015; Munir, 2016; Xu et al., 2016). A higher ratio, for instance, an elevated PM\textsubscript{2.5} contribution, is usually suggestive of high levels of secondary aerosols such as nitrates,
sulphates and ammonium while a low ratio is representative of dust from on-road vehicles, wind-blown dust as well as desert sand (Brook et al., 1997; Chan and Yao, 2008; Wang et al., 2014). The study of ratio is hence used to determine whether an emission originates from an anthropogenic source, where the ratio is high, or a natural crustal source, where the ratio is low (Brook et al., 1997; Xu et al., 2016).

Meteorological factors can strongly influence the quality of ambient air through complex interactions between different processes of particulate matter such as transport, emissions, chemical transformation, as well as wet and dry deposition (Seinfeld and Pandis, 1998; Demuzere et al., 2009). Locally, in addition to some chemical processes, biogenic and dust emissions are regulated by local weather elements, such as relative humidity, temperature, solar radiation and cloudiness. Regionally, the short and long-range transport of particulate matter relies on the characteristics of the boundary layer turbulence and synoptic atmospheric circulation (Demuzere et al., 2009; Juneng et al., 2011; Tuygun and Elbir, 2020). In Malaysia, the association between local and regional weather elements with PM$_{10}$ is commonly reported in studies (Juneng et al., 2011; Dominick et al., 2012b; Afzali et al., 2014; Latif et al., 2014; Rahman et al., 2015; Hassan et al., 2020). A study by Juneng et al. (2011) showed that the three factors influencing PM$_{10}$ variations are local meteorological aspects, conditions of synoptic weather and the number of fire hotspots that represent regional biomass emissions.

The reported research on PM$_{2.5}$ in Malaysia has covered both qualitative and quantitative studies undertaken at different background areas, predominantly on the Malaysian Peninsula. A sampling site in the urban-industrial area of the Klang Valley, which is one of the Global Atmosphere Watch (GAW), has attracted the interest of researchers studying the chemical composition of PM$_{2.5}$,
particularly when influenced by Indonesian peatland fires. A study by Amil et al. (2016) in the urban-industrial area of the Klang Valley showed that 43% of the PM$_{2.5}$ daily mean concentration exceeded the World Health Organisation’s (WHO) guideline value given at the time. Since the middle of 2017 though, the Malaysian Department of Environment has established continuous monitoring of fine particulate matter (or PM$_{2.5}$) at 65 stations throughout Malaysia. Therefore, this study aims to investigate the level of PM$_{2.5}$ and its spatiotemporal variations and compare them to PM$_{10}$. This study also investigates the correlation of both PM$_{2.5}$ and PM$_{10}$ to other major air pollutants such as carbon monoxide (CO), ozone (O$_3$), nitrogen dioxide (NO$_2$) and sulphur dioxide (SO$_2$) as well as meteorological parameters. The Principal Component Analysis (PCA) method, together with Kaiser-Meyer-Olkin (KMO), was used to study the associations based on the pattern of correlation between particulate matter and other gases and meteorological parameters recorded at each station.

2 METHODOLOGY

2.1 Study Area Description
Malaysia has tropical weather with two monsoon seasons, namely the Northeast Monsoon (NEM, December to February) and the Southwest Monsoon (SWM, June to August) with the Intermonsoon (ITM) seasons in between (Tangang et al., 2008; Ngai et al., 2017; Tangang et al., 2017; Jamaluddin et al., 2018; Hassan et al., 2020). However, in 2018, MetMalaysia reported that the NEM occurred from November to March and SWM occurred from May to September. Malaysia consists of two parts which are separated by the South China Sea. These are the Malaysian Peninsula and East Malaysia (otherwise known as Malaysian Borneo). Across these two parts are located 65 continuous air quality monitoring stations which are run by the Malaysia Department of
Environment (DOE). These stations are divided into four regions in the Malaysian Peninsula; northern, central, southern and eastern, and two regions in East Malaysia; Sabah and Sarawak. These stations are also categorized as urban, suburban, rural or industrial. The location of the stations and the geography of Malaysia is illustrated in Fig. 1 and Table S1.

[Fig. 1]

The northern part of the Malaysian Peninsula is less developed. It mainly consists of agriculture land, particularly the production of rice and sugarcane, as the main economic activity. The centre and lower parts of the northern region are more developed with commercial business and industrial activities (DOSM, 2017). The central part of the Malaysian Peninsula is the most developed region in the country and is where the capital city, Kuala Lumpur, is located. According to the DOSM (2018) in the year 2018, the central region was the most densely populated with an estimated population of approximately 9.5 million. The southern region has a rather flat landscape while the northern part of this region has a slight elevation from the peak of Titiwangsa Ridge. The main economic activity in this region is manufacturing, in addition to palm oil plantations, fisheries and wood-based industries (DOSM, 2017). The eastern region is less developed compared to the western region. In the eastern region, fisheries are the main economic activity (DOSM, 2017). The East Malaysia consists of Sabah and Sarawak on the island of Borneo. When it comes to economic activity, these regions are less developed but rich in palm oil plantations and wood-based industries, as well as popular ecotourism destinations (DOSM, 2017).

2.2 Data Collection

2.2.1 Air quality data
The hourly data for PM$_{2.5}$ and PM$_{10}$ as well as other gas air pollutants such as CO, O$_3$, SO$_2$ and NO$_2$ were retrieved from the DOE Malaysia between January 2018 and December 2018. The hourly PM$_{10}$ and PM$_{2.5}$ measurements presented in this study were retrieved using a 1405-DF Tapered Element Oscillating Microbalance (TEOM™) Continuous Dichotomous Ambient Air Monitor which allows for the real time measuring of the mass concentrations of PM$_{2.5}$ and PM$_{10}$. Hourly data for CO, O$_3$, SO$_2$ and NO$_2$ were measured using Thermo Scientific 48i (USA), 49i (USA), 43i (USA) and 42i (USA), respectively. The air pollutants data went through quality assurance and quality control (QA/QC) procedures before being submitted to the DOE.

2.2.2 Meteorological data

The meteorological data (relative humidity, temperature, wind speed, and solar radiation) from DOE was recorded using a Climatronics AIO 2 Weather Sensor by the Climatronics Corporation, USA. As for precipitation data, satellite data (Global Precipitation Measurement, GPM) from NASA was used to study the influence of precipitation on air quality in different regions of Malaysia. The GPM data had a spatial scale of 0.1° and a temporal scale of 30 min. It was downloaded and analysed for the year 2018 and for Malaysia alone. In Malaysia’s tropical region, precipitation consists of only rainfall hence in this text, hereafter, the term ‘rainfall’ is used to further discuss the association of rainfall with air quality.

2.3 Data Analysis

As this study will focus on the different regions of Malaysia, data from all 65 stations in respective regions were analysed to represent the results of each region. The hourly data for PM$_{2.5}$ and PM$_{10}$ were calculated so as to study the overview of diurnal and seasonal patterns of PM$_{2.5}$ and PM$_{10}$ as well as the ratio of PM$_{2.5}$ to PM$_{10}$. The statistical method used in this study included descriptive
analysis such as mean, minimum, maximum, standard deviation and Spearman’s Rank Correlation. The Spearman’s Rank Correlation test is used to measure the association between two variables which in this study are pollutants and meteorological aspects.

In addition to the above, multivariate analysis, such as Principal Component Analysis (PCA), was employed in this study using hourly data. The PCA method is used to summarize a set of variables based on their characteristics. It does this by transforming a set of interrelated variables into uncorrelated variables. The transformed variables (also known as principal components, PCs) are correlated to a linear combination of the original set of variables (Abdul-Wahab et al., 2005; Viana et al., 2006; Škrbić and Durišić-Mladenović, 2007; Sousa et al., 2007; Juneng et al., 2009; Dominick et al., 2012a).

A better relationship between the PCs and the original set of data can be obtained by applying PC varimax rotation. This ensures that each variable is optimally correlated with only one PC and has zero correlation with other PCs (Statheropoulos et al., 1998). Varimax factors (VFs), also known as factor loading, are obtained by running PC varimax rotation with PCs that show eigenvalues greater or equal to 1 (Kim et al., 1978). Variables that have a high factor loading have a high contribution to the variation accounted for by the specific PC (Jolliffe, 1986). Factor loading values which are greater than 0.75 are considered as “strong” (Liu et al., 2003; Azid et al., 2014). In this study, any factor loadings over 0.70 were selected for results interpretation.

Principal components can be expressed as in Eq. 1 below:

$$Z_{ij} = a_{i1}X_{1j} + a_{i2}X_{2j} + \ldots \ldots + a_{in}X_{nj} \quad \text{(Eq. 1)}$$
Where, $Z_{ij}$ is the component score for component $i$ and for sample $j$, $a$ is the component loading and $x$ is the measured value of the variable while “$n$” is the total of the variables. The Kaiser-Meyer-Olkin (KMO) test also measures the suitability and adequacy of the data sets for PCA and will be acceptable if the value of KMO is greater than 0.5 (Field, 2013).

### 3 RESULTS AND DISCUSSION

#### 3.1 Descriptive Analysis of PM$_{2.5}$ and PM$_{10}$

The descriptive analysis of PM$_{2.5}$ and PM$_{10}$ were compared in six respective regions of Malaysia. Table S2 depicted the overall 24-h mean of PM$_{2.5}$ and PM$_{10}$ of each station in the respective region while Fig. 2 presented the mean of hourly data based on each region. The mean concentrations of PM$_{2.5}$, as seen in Fig. 2 and Table S2, show that the highest mean concentration of PM$_{2.5}$ ($23 \pm 8 \mu g/m^3$, range = 4.6 – 158 $\mu g/m^3$) in Malaysia was recorded in the central region and the lowest mean concentration in the Sabah region ($11 \pm 5 \mu g/m^3$, range = 1.1 – 60 $\mu g/m^3$). In contrast to the central region and other regions in Malaysia, the Sabah region is a pristine area. Besides wood-based industry and palm oil plantations, the economic activities in the Sabah region involve ecotourism in the national parks and national marine parks. Contrarily, the central region itself is the economic hub of the country, with a high-density population that is estimated to be approximately 9.5 million (DOSM, 2018) and has high volume of traffic as compared to other regions (MOT, 2019). Therefore, the concentration of PMs is generally high in this region. The study by Ab. Rahman et al. (2022) also denote that Klang Valley is a highly polluted area in Malaysia.

![Fig. 2]
In this study, the mean concentrations of PM$_{2.5}$ for the northern, southern and eastern regions on the Malaysian Peninsula are $17 \pm 8 \ \mu g/m^3$ (range = $2.1 - 120 \ \mu g/m^3$), $17 \pm 7 \ \mu g/m^3$ (range = $0.8 - 112 \ \mu g/m^3$) and $16 \pm 8 \ \mu g/m^3$ (range = $1.7 - 84 \ \mu g/m^3$), respectively. The Sarawak region recorded the second lowest mean concentration of PM$_{2.5}$ of $13 \pm 9 \ \mu g/m^3$ (range = $1.8 - 178 \ \mu g/m^3$). The Sarawak region on the island of Borneo which has an approximate of 2.8 million population (DOSM, 2018) is less developed compared to the Malaysian Peninsula thus has a lower concentration of PM$_{2.5}$. Similar with the Sabah region, the main economic activities in the Sarawak region are palm oil plantations and wood-based industries besides ecotourism with several national parks (DOSM, 2017). The economic activities undertaken in the northern and southern regions of the Malaysian Peninsula vary considerably. In the northern region, for example, agriculture, commercial business and industrial activities are common whereas the southern region is known for manufacturing, palm oil plantations, fisheries and wood-based industries (DOSM, 2017). The eastern region recorded the lowest concentration of PM$_{2.5}$ in the Malaysian Peninsula which can be explained as this region is the least commercially developed compared to other region in Peninsular Malaysia (DOSM, 2017). The western part of this region is a mountainous area which most of it are national parks.

As for the mean concentration of PM$_{10}$ in Fig. 2 and Table S2, the central region indicates the highest mean concentration of PM$_{10}$ ($32 \pm 10 \ \mu g/m^3$, range = $6.0 - 181 \ \mu g/m^3$) throughout Malaysia, the same is demonstrated by the PM$_{2.5}$ concentrations. The emissions from shipping (Pandolfi et al., 2011; Bove et al., 2016; Ledoux et al., 2018; Wu et al., 2020), industrial activities (Karar et al., 2006; Park and Dam, 2008; Jamhari et al., 2014; ChooChuay et al., 2020; Flores et al., 2020) and mobile sources (Negral et al., 2008; Kulshrestha et al., 2009; Khan et al., 2015;
Azhari et al., 2020; ChooChuay et al., 2020) undoubtedly contribute to the higher concentrations of PMs. For instance, CA21B station, which is located in the Klang Valley area, an urban site near to the country’s busiest shipping port - Port Klang, frequently has the highest concentration of PM10 (Rahman et al., 2015; Mohtar et al., 2018). Port Klang is the central logistical hub of Malaysia and one of the busiest ports in the world and Southeast Asia (Soon and Lam, 2013; Mustaffa et al., 2016).

In addition, the mean 24-h concentrations of PM$_{2.5}$ and PM$_{10}$ were compared to the Malaysian Ambient Air Quality Standard (MAAQS) Interim II (2018). The MAAQS Interim II states that the concentration limit for 24-h averaged PM$_{2.5}$ and PM$_{10}$ are 50 µg/m$^3$ and 120 µg/m$^3$, respectively, while, the concentration limits for annual averaged PM$_{2.5}$ and PM$_{10}$ are 25 µg/m$^3$ and 45 µg/m$^3$, respectively. Overall, the average concentration of PM$_{2.5}$ recorded in Malaysia is lower than the value suggested for the 24-h mean concentration in the MAAQS Interim II guideline.

### 3.2 Diurnal and Seasonal Variations of PM$_{2.5}$ and PM$_{10}$

The concentration of air pollutants in the lower atmosphere tends to exhibit distinct diurnal variations as the most intense human activity tends to occur during day time as well as the atmospheric mixing height due to solar heating. Diurnal variations of PM$_{2.5}$ and PM$_{10}$ are presented in Fig. 3 (a) to Fig. 3 (f). It was found that the central region generally had the highest daily mean concentrations for all pollutants (Fig. 3 (b)) whilst the lowest concentrations were measured in the Sabah region (Fig. 3 (e)). From Fig. 3, it can be seen that the diurnal pattern for PM$_{2.5}$ across Malaysia is the same and in a bimodal pattern. The first PM$_{2.5}$ and PM$_{10}$ concentration peak observed occurred in the morning while the second one was at night.
The peak in the morning indicates that the emission of particulate matter originates from motor vehicle usage by people performing their morning routine of heading out to work and school (Kuttler and Strassburger, 1999; Wang and Christopher, 2003; Jamil et al., 2011; Yoo et al., 2015). The lower concentrations of PM\textsubscript{2.5} and PM\textsubscript{10} in the afternoon, from 1:00 pm to 4:00 pm local time, can be explained by a lower level of outdoor activities and hence lower vehicle emissions (Norazian et al., 2015). Additionally, the reduction of PM\textsubscript{2.5} and PM\textsubscript{10} could also result from the increased mixing height lowering the concentrations of both PM\textsubscript{2.5} and PM\textsubscript{10} (Ulke and Mazzeo, 1998; Lal et al., 2000; Langner et al., 2011; Hassan et al., 2020; Huang et al., 2020; Liu et al., 2021). As for the second peak at night, this could be attributed to the evening rush hour and lower wind speed which results in the slower transportation of PM away from the test area. Besides, the mixing layer begins to get lower in the late evening as the atmosphere starts to cool down, which accounts for the higher concentration of PM at night. However, the concentration of PM recorded in Sabah reduced significantly from 9:00 pm local time. Based on our investigation, most of the air quality monitoring stations in Sabah are located near the sea. It is speculated that the stability condition during night time may be reduced by the land breeze (Ooi et al., 2017). Meteorological conditions, such as atmospheric stability (Juneng et al., 2011; Ooi et al., 2017) and wind speed (Afroz et al., 2003; Radzi Bin Abas et al., 2004), influence the peak concentration of PM in the late evening to night.

The particles which are suspended in the atmosphere travel with the movement of air. Thus, the different monsoon seasons experienced in Malaysia can lead to seasonal fluctuations of PM\textsubscript{2.5} and PM\textsubscript{10}. Generally, there are two seasonal variations which are the Northeast Monsoon (NEM), Southwest Monsoon (SWM) and separated by the Inter-Monsoon (ITM) season (MetMalaysia,
The solid red box in Fig. 3 (g) until Fig. 3 (l) indicates the NEM (November to March) in Malaysia and the red dashed line box indicates the SWM (May to September). In general, the highest monthly concentrations of PM$_{2.5}$ and PM$_{10}$ were consistently recorded in the central (Fig. 3 (h)) region with a PM$_{2.5}$ range from 18 to 29 µg/m$^3$ and a PM$_{10}$ range from 26 to 40 µg/m$^3$. The lowest monthly concentrations of particulates were in the Sabah (Fig. 3 (k)) region where a range of 5 to 18 µg/m$^3$ for PM$_{2.5}$ concentrations and 15 to 27 µg/m$^3$ for PM$_{10}$ concentrations were recorded.

Based on Figs. 3g-l, the seasonal pattern for PM$_{2.5}$ and PM$_{10}$ is identical for all regions, i.e., when the concentration of PM$_{10}$ is high, so too is the concentration of PM$_{2.5}$. The SWM season demonstrated the highest concentration of PM$_{2.5}$ and PM$_{10}$ throughout the year. During SWM, the dry season occurrence is frequent where the highest count of local, as well as regional, biomass burning occurs (Heil and Goldammer, 2001; Keywood et al., 2003; Tangang et al., 2010; Khan et al., 2015; Kusumaningtyas and Aldrian, 2016; Shannon et al., 2016). Also, the predominantly low-level southwest winds, which dominate the region during this season, have been found to be responsible for transporting these pollutants to Malaysia mainly from Sumatra Island, Indonesia (Juneng et al., 2009; Khan et al., 2015; Lee et al., 2017). Coupled with the fact that haze episodes occurred in August 2018 mainly in the central and northern regions of Peninsular Malaysia as well as in Sarawak, East Malaysia, the highest concentrations of PM$_{2.5}$ and PM$_{10}$ were also recorded in that month (Kawi, 2018; NSTOnline, 2018; Nufael, 2018; MetMalaysia, 2019; ASMC, 2022b, a).

Overall, throughout the NEM season, the concentrations of PM$_{2.5}$ and PM$_{10}$ fluctuated between January and March and then November until December for all regions. Solar radiation and surface temperature (Fig. S4) both increased indicating the dry season in February and March leading to a high number of local biomass burning events which in turn caused a rise in PM concentrations (Dominick et al., 2015). The ITM season recorded a declining concentration for both PM$_{2.5}$ and
PM$_{10}$ in April and October for all regions. This is believed to be due to a weaker wind with no clear
direction lowering concentrations (Keywood et al., 2003; Mohd Akhir et al., 2014; Ooi et al., 2017;
Lolli et al., 2019).

3.3 Influence of Other Gases and Meteorological Factors

The association of PM concentrations with meteorological factors (temperature, wind speed,
relative humidity, rainfall and solar radiation) and other criteria pollutants (SO$_2$, NO$_2$, O$_3$, CO) is
depicted by the Spearman’s Rank Correlation Coefficient ($r$) in Table 1 and Table S5a-c, and the
PCA analysis summarised in Table 2 and Table S6a-c. In all regions, PM$_{10}$ and PM$_{2.5}$
concentrations were found to be significantly associated ($p < 0.01$) with concentrations of other gas pollutant$s$ such as NO$_2$ and CO. Moderately positive correlations were observed in the association
between PM$_{10}$ (PM$_{2.5}$) and NO$_2$ in the northern, central and southern regions with $r = 0.41$ (0.42),
$r = 0.48$ (0.47) and $r = 0.41$ (0.41) respectively. As for the correlation of PM$_{10}$ (PM$_{2.5}$) to CO,
moderately positive associations were recorded in the northern, central and eastern regions with $r$
= 0.43 (0.44), 0.48 (0.47) and 0.45 (0.45) respectively. From the positive correlations observed
between PM with NO$_2$ and CO, it is possible that the PM in the region is emitted simultaneously
with NO$_2$ and CO, to which it was closely related to, through combustion processes which include
combustion from motor vehicles (Dominick et al., 2012b).

[Table 1]

[Table 2]

Following on from the previous paragraphs on seasonal variations, the concentrations of particulate
matter were observed to be high when the temperature was high, there was high solar radiation,
low relative humidity and low rainfall (Fig. S4). The high concentrations of particulate matter
observed during a low rainfall were caused by the direct effect of rainfall as the washing effect
decreases the concentration of pollutants (Kwak et al., 2017; Gao et al., 2019). Hence when there is little rainfall, the washing effect does not occur thus causing PM concentrations to be higher. However, a further analysis of correlations (shown in Table 1 as well as Table S5a-5c) demonstrated a significantly weak correlation \( p < 0.01 \) for all meteorological parameters, such as local wind speed, relative humidity, solar radiation, temperature and rainfall. This applied to all regions in Malaysia during the study period.

The PCA results for all the different regions in Malaysia produced two factors during the annual cycle and three factors during seasonal variations with eigenvalues greater than 1. These were used in PCA interpretation (Table 2 and Table S6a-c) for the possible emission sources and/or conditions influencing air pollutants’ concentrations. The first factor (F1) showed a strong negative factor loading with relative humidity (RH) and a strong positive factor loading with ambient temperature (Temp) and O\(_3\) for all regions. It is indicated that meteorological conditions, such as temperature as well as relative humidity, strongly influence the efficiency of photochemical processes leading to ozone formation and influencing the formation of secondary aerosols which form particulate matter (Li et al., 2017; Mao et al., 2020). Higher temperatures and lower relative humidity are conducive to O\(_3\) formation during ozone episodes (Yang et al., 2020) which can be easily formed in these conditions even though O\(_3\) pollution is more dependent on vehicle emissions (Zhao et al., 2018).

The second factor (F2) presented a strong positive factor loading with PM\(_{10}\) and PM\(_{2.5}\) for all regions whereas CO and NO\(_2\) showed positive factor loadings for all regions apart from Sabah and Sarawak. This observation can be explained by the location of each region. The northern, central, southern and eastern regions are located on the Malaysian Peninsula whereas the Sabah and
Sarawak regions are located on the island of Borneo. The Malaysian Peninsula has experienced more expansion, urbanisation, as well as economic development compared to the island of Borneo. These changes have resulted in rise of anthropogenic emissions, particularly from combustion-related activities on the Malaysian Peninsula, which contribute to the formation of NO\textsubscript{2} and CO, along with particulate matter (Azhari et al., 2018; Halim et al., 2020; Latif et al., 2021).

During the NEM and SWM season (Table S6a-6b), the third factor (F3) observed a strong positive correlation between SO\textsubscript{2} and NO\textsubscript{2}, particularly in the central region, for both NEM and SWM. The sources of SO\textsubscript{2} in the region are predominantly emissions from power stations in the central region. These use resources such as coal, gas, steam and biomass (Baruya, 2010; Salahudin et al., 2013). Besides emissions from power stations, industrial sectors as well as heavy-duty vehicle emissions contributed to the higher concentration of SO\textsubscript{2} (Pereira et al., 2007; Mohamad et al., 2015). Meanwhile, NO\textsubscript{2} levels in the central region were mainly influenced by motor vehicles, followed by power stations and industrial sector emissions (Azmi et al., 2010; Salahudin et al., 2013; Latif et al., 2014; Mohtar et al., 2018; Wang et al., 2020). It can be concluded that the third factor (F3) from PCA originated from the emission of anthropogenic activities in the region which influenced the concentrations of SO\textsubscript{2} and NO\textsubscript{2}.

### 3.4 Ratios of PM\textsubscript{2.5} to PM\textsubscript{10}

The overview of spatial ratios of PM\textsubscript{2.5} to PM\textsubscript{10} is presented in Fig. 4 and Table S2. When comparing the contribution of each region towards the ratio of PM\textsubscript{2.5} to PM\textsubscript{10}, the central region demonstrated the highest mean ratio of PM\textsubscript{2.5} to PM\textsubscript{10} (0.72, range = 0.68 – 0.76). For the northern and southern regions, the mean ratio of PM\textsubscript{2.5} to PM\textsubscript{10} was 0.66 (range = 0.56 – 0.73) and 0.66 (range = 0.57 – 0.75), respectively. The mean ratio of PM\textsubscript{2.5} to PM\textsubscript{10} in the eastern region was
recorded as 0.64 (range = 0.58 – 0.68). The lowest mean ratio of PM$_{2.5}$ to PM$_{10}$ nationwide was recorded in Sabah, followed by Sarawak with a value of 0.56 (range = 0.51 – 0.67) and 0.60 (range = 0.54 – 0.68), respectively.

The diurnal mean ratios of PM$_{2.5}$ to PM$_{10}$ for different regions are depicted in Fig. 5 (a-f). Overall, the highest mean diurnal ratio of PM$_{2.5}$ to PM$_{10}$ was recorded by the central region and the lowest recorded by Sabah. All regions in Malaysia that recorded the morning peak PM concentrations showed lower PM$_{2.5}$ to PM$_{10}$ ratios compared to the night peak which exhibited a slightly higher ratio of PM$_{2.5}$ to PM$_{10}$. This indicates that the emissions of PM in the morning were dominated by coarse particulate matter contribution rather than fine particulate matter while the emissions of PM at night were dominated by fine particulate matter contribution rather than coarse particulate matter. The lowest ratio for all regions was recorded between 7:00 am to 8:00 am. This might be due to resuspended coarse particulate matter from road dust and relate to the starting time of daily human activities (Munir et al., 2013; Xu et al., 2016; Coskuner et al., 2018).

The annual cycle of PM$_{2.5}$ to PM$_{10}$ ratios is further discussed according to the annual cycle of PM in different regions based on Fig. 3 (g-l). In Fig. 5 (g-l), the NEM is indicated by the solid box and the SWM is indicated by the dashed line box. The highest concentrations of PM$_{2.5}$ and PM$_{10}$ observed during the SWM for all regions (Figs. 3 (g-l)) also show the highest ratio of PM$_{2.5}$ to PM$_{10}$ across Malaysia. The same pattern of PM$_{2.5}$ to PM$_{10}$ ratios in Malaysian Borneo (Sabah and Sarawak) with a peak in August, which is the haze period, indicates that the contribution of PMs is mainly from PM$_{2.5}$. This can be seen in the Malaysian Peninsula regions as well which show a
higher PM$_{2.5}$ to PM$_{10}$ ratio being recorded during the SWM. This can be explained by the transboundary movement of PM$_{2.5}$ where the fine particulate matter is more easily transported to Malaysia than the coarse particulate matter (PM$_{10}$). A higher PM$_{2.5}$ concentration leads, in effect, to a higher ratio of PM$_{2.5}$ to PM$_{10}$. The study by Gao and Ji (2018) also recorded higher PM$_{2.5}$ to PM$_{10}$ ratios during the haze period.

In general, the ratios of PM$_{2.5}$ to PM$_{10}$ recorded for each region were generally consistent in terms of patterns throughout the days and months of the study (Fig. 5). From Fig. 4, however, note that the ratio of PM$_{2.5}$ to PM$_{10}$ for the central region was higher (0.72, range = 0.68 to 0.75) than other regions (Table S2). The fact that the ratios of PM$_{2.5}$ to PM$_{10}$ in the central region were often high indicates two possibilities. Firstly, that the central region potentially constitutes a high magnitude of emissions which tended to release much greater quantities of fine particles rather than coarse particles. The central region is the most densely populated part of the country and is rapidly developing through industrialisation. Therefore, it is likely that industries, surface transportation and burning activities act as the main sources of fine particulate matter (Amil et al., 2016; Sulong et al., 2017). Secondly, the abundance of PM$_{2.5}$ and PM$_{10}$, and their ratios, are dependent on the wind profile and the distance of the monitoring sites from the sources (Vu et al., 2015). Sites that are closer to the sources of PM will consequently have much higher levels and ratios of the measured pollutants. The abundance of monitoring stations in the central region, which is located within the urbanised and industrial areas, also influences the ratios of PM and as this study is averaging PM over the region, this will also contribute to higher ratios of PM$_{2.5}$ to PM$_{10}$. Overall, the results of our ratio analysis have provided evidence of the higher abundance of fine particulates to coarse particulates in the ambient air.
4 CONCLUSION

This study focused on the spatiotemporal variations of PM$_{2.5}$ and PM$_{10}$ in Malaysia and their relationship with other criteria pollutants and meteorological aspects. Overall, the study revealed that the central region is the most polluted area in Malaysia with the 24-h mean reading for PM$_{2.5}$ being $23 \pm 8 \, \mu g/m^3$ and ranging from $4.6 \, \mu g/m^3$ to $158 \, \mu g/m^3$ and the 24-h mean for PM$_{10}$ being $32 \pm 10 \, \mu g/m^3$ with a range from $6.0 \, \mu g/m^3$ to $181 \, \mu g/m^3$. Regarding the bimodal pattern of diurnal concentrations observed in all regions, the central region recorded the highest diurnal concentration while the Sabah region recorded the lowest. The first peak in the morning indicates that PM emissions originate from motor vehicles while the second peak, occurring in the night, relates to the accumulation of PM as well as meteorological conditions such as atmospheric stability.

Seasonal fluctuations of PM$_{2.5}$ and PM$_{10}$ indicate that the month of August, which is during the southwest monsoon period (dry season) in Malaysia, recorded the highest reading of PM in the central region and the lowest in the Sabah region.

Further quantitative analysis observed weak to moderately positive correlations between PM and NO$_2$ (ranges from 0.29 to 0.48) as well as CO (ranges from 0.23 to 0.48) in all regions. The second factor loading for both of these gases indicated a strong positive loading in PCA for all regions apart from Sabah and Sarawak, which are the least urbanised regions. This shows that the main pollutants on the Malaysian Peninsula, besides PM, are CO and NO$_2$. The first factor loading indicated a strong negative factor loading with relative humidity and a strong positive factor loading with temperature and O$_3$ for all regions, indicating the potential of secondary aerosols formation. The ratios of PM$_{2.5}$ to PM$_{10}$ showed that the central region recorded the highest mean ratio (0.72) while the lowest mean ratio (0.56) was recorded in the Sabah region. These results indicate that there was a higher abundance of fine particulates than coarse particulates in the
ambient air, which in effect correlates to their being a greater risk posed to human health. In conclusion, this in-depth study of the spatiotemporal distribution of PM$_{2.5}$ in Malaysia has suggested that PM$_{2.5}$ is predominantly concentrated over regions where heavy anthropogenic activities occur and thus implies the potential health risks threatening the populations of these areas. As the suggestion for future studies, it will be good to study the distribution and association of criteria pollutants (SO$_2$, NO$_2$, O$_3$, CO) along with PM$_{2.5}$ and PM$_{10}$ to estimate pollutant sources in Malaysia.

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Ethics approval and consent to participate

Not applicable

Consent to publish

Not applicable

Competing interests

The authors declare that they have no competing interests.
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Authors' contributions

Anis Asma Ahmad Mohtar: conceptualization, formal analysis, investigation, visualization, writing original draft, reviewing and editing; Mohd Talib Latif: conceptualization, funding acquisition, supervision, writing, reviewing and editing; Doreena Dominick: formal analysis, investigation, visualization; Maggie Chel Gee Ooi: writing, reviewing and editing; Azliyana Azhari: writing, reviewing and editing; Nor Hafizah Baharudin: writing, reviewing and editing; Norfazrin Mohd Hanif: writing, reviewing and editing; Jing Xiang Chung: formal analysis, visualization; Liew Juneng: supervision, writing, reviewing and editing.

Availability of data and materials

The datasets analysed during the current study are not publicly available and belong to the Malaysian Department of Environment. The data is available from the corresponding author on reasonable request.


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Table 1 The Spearman’s rank correlation of particulate matters, gases and meteorological parameter based on different region in Malaysia

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Northern PM₁₀</th>
<th>PM₂.₅</th>
<th>PM₁₀</th>
<th>PM₂.₅</th>
<th>Southern PM₁₀</th>
<th>PM₂.₅</th>
<th>PM₁₀</th>
<th>PM₂.₅</th>
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<th>Sabah PM₁₀</th>
<th>PM₂.₅</th>
<th>Sarawak PM₁₀</th>
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<td>0.93**</td>
<td>1.00</td>
<td>0.93**</td>
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<td>0.41**</td>
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** Asterisk shows the correlation significant at the 0.01 level (2-tailed)
* Bold numbers indicate strong correlation between parameters
Table 2 The Principal Component Analysis (PCA) results of PM, criteria gases and meteorological parameter based on region.

<table>
<thead>
<tr>
<th>Region</th>
<th>Northern</th>
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* Bold numbers indicate strong factor loading and italic numbers indicate moderate factor loading
Fig. 1 The station location of particulate matter sampling sites over Malaysia separated into six regions, namely northern, central, eastern, southern in Peninsular Malaysia and Sabah and Sarawak in East Malaysia.
Fig. 2 The box plot of (a) PM$_{2.5}$ and (b) PM$_{10}$ concentration based on the mean of hourly data in all stations of each region over Malaysia.
Fig. 3 The diurnal (left panel) and monthly (right panel) mean concentration of PM$_{2.5}$ and PM$_{10}$ based on different region in Malaysia. On the right panel, the red solid box indicates the northeast monsoon (NEM) and the red dashes box indicates the southwest monsoon (SWM) in Malaysia.
**Fig. 4** The spatial distribution of PM$_{2.5}$ to PM$_{10}$ ratios over six regions in Malaysia which indicated by different symbol in each region.
Fig. 5 The diurnal (left panel) and monthly (right panel) mean ratio of PM$_{2.5}$ to PM$_{10}$ at different regions in Malaysia. On the right panel, the solid box indicates the northeast monsoon (NEM) and the dashes box indicates the southwest monsoon (SWM) in Malaysia.