DIURNAL TREND OF PARTICULATE MATTER CONCENTRATION AT INDUSTRIAL AREA USING MULTIVARIATE ANALYSIS

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Abstract: Particulate matter is a dominant air pollutant in Malaysia and it is a major emission that has caused many respiratory problems. In this study, the influence of trace gases and meteorological parameters with particulate matter that has an aerodynamic diameter of less than 10 μ m (PM₁₀) were investigated. Simple linear regression was used to analyse the strength of trace gases and meteorological parameters with PM₁₀, while the Principal Component Analysis (PCA) was used to determine the sources of air pollution. The average concentration of PM₁₀ for a year (January 2010 to December 2010) in Pasir Gudang was 56.567 µg/m³, which exceeded the New Ambient Air Quality Standards and international guidelines. The highest mean daily was recorded at 9 a.m (LT) (63.644 µg/m³), while the lowest was at 11 p.m. (LT) (31.44 µg/m³) (12-60 µg/m³). PM₁₀ concentrations are significantly correlated with SO₂ (R²=0.0302, p<0.01), CH₄ (R²=0.0817, p<0.01), RH (R²= 0.0581, p<0.01), NO_x (R²=0.1118, p<0.01) and NO (R²=0.0786, p<0.01). The major sources of PM₁₀ are traffic combustion at 35.505%, followed by meteorological factors at 28.972% and traces gases from industry, such as SO, and NO_x, at 11.009%.

Keywords: Particulate matter, ambient air, principal component analysis, simple linear regression, industrial area.

Introduction

One of the major issues today is air pollution, affects which human health and the environment. Air pollution can occur in the form of suspended particulates, liquid or gas in the air, which are present in the atmosphere in sufficient concentration in the building or outdoor environment (Latif et al., 2018). These scenarios create quantifiable impacts on humans, as well as other creatures such as animals, vegetation and materials (Branco et al., 2019). Emission and transmission of air pollutants into the atmosphere directly increase the ambient concentration (Winkler et al., 2018). Two major sources that contribute to the pollution of ambient air are anthropogenic and natural sources (Abdullah et al., 2016; Cao et al., 2019). Nitrogen oxides (NO), particulate matter with an aerodynamic diameter less than $10 \ \mu\text{m} (\text{PM}_{10})$, sulphur oxides (SO_x), ozone (O₃), Carbon monoxide (CO) and hydrocarbons (HC) are mainly outdoor air pollutants. Aerosols (also known as particulate matter or PM) are one of the most harmful air pollutants as they have caused more than 7 million premature deaths annually worldwide (WHO, 2015; Sanchez-soberon *et al.*, 2019). PM consists of a mixture of airborne solid particles, liquids droplets, and it comprises an array of different materials, such as sea spray, fly ashes, pollen and soot (Chamseddine *et al.*, 2019).

In Malaysia, mobile sources, such as emissions from motor vehicles, or stationary sources, like emissions from factories, power plants and open burnings are the major contributors to air pollution. Air pollution in urban and industrial areas is associated with a range of health effects, such as cardiovascular and respiratory problems (Vardoulakis et al., 2015; Peixoto et al., 2017; Chamseddine et al., 2019). Khailili et al. (2018), in their study, examined traffic-related pollutants and concluded that the major contributors especially for PM₁₀ and PM_{2,5}, were motor vehicles. Particulate matter can come from exhaust and non-exhaust soruces. Non-exhaust sources include particles from the road surface, dust resuspension, tire wear, and brakes (Kalimeri et al., 2019). Particulate matter (PM_{10}) was found to have an effect on paediatric asthma attack, especially towards sensitive groups, such as young and older people, as well as people suffering from respiratory problems (Stamatelopulou et al., 2019).

Many studies have shown that exposure to PM_{10} 150 m to 300 m from the road poses a high risk towards the cardiovascular system (Das et al., 2018; Schibuola et al., 2019; Stamatelopulou et al., 2019). This is because the small size of the particulate matter, which is less than 10 µm, is easily inhaled and can penetrate respiratory airways. Common chronic lung diseases faced by the sensitive group exposed to PM₁₀ is asthma caused by the airways becoming inflamed and increases in the production of mucus, which cause difficulty in air flowing in and out of the lung. Asthma exacerbation risk are caused by several environmental exposures, and one of the ambient air pollutants that plays an important role in adverse respiratory outcomes was PM₂₅ - PM₁₀ (Meng *et al.*, 2010; Kang *et al.*, 2019).

The concentration of ozone precursors shows a slight variation during a specific time caused by the concentration of ozone in the atmosphere (Khan *et al.*, 2015). Ozone is a primary component in photochemical smog, with radiative forcing of 0.4 ± 0.20 W/m² (Pawlak & Jaroslawki, 2019). The photolysis process of ozone is the dominant source of hydroxyl radicals (OH), which act as primary oxidant. Thus, the degradation of trace gases in the atmosphere is dominated by the reaction of trace gases with OH, and there is a significant number of halocarbons, CO and CH₄ (Fang *et al.*, 2018). OH can remove almost all pollution, such as

volatile organic matter (VOC), and carbon monoxide (CO) is a combination of both of these pollutants and will produce hydroperoxy (HO₂) and peroxy (RO_2) radicals. The presence of NO in the air causes a reaction with RO₂, which can produce HO, and OH radicals reformed by the reaction between HO₂ and NO with NO₂ as a by-product that degrades photochemically to produce ozone (Awang et al., 2015; Fang et al., 2018; Oh et al., 2019). A combination of nitrous oxides in the atmosphere can cause paralysis of the nervous system and death in animals and humans at a concentration of 100 µg/m3 (Mohamed et al., 2016; Chamseddine et al., 2019). It is important to look at the trend of air pollutants in the risk of ambient air to prevent long-term effects. The objectives of this study are to investigate the diurnal trend of PM_{10} , as well as to quantify the air pollution sources in Pasir Gudang via Principal Component Analysis (PCA) to provide beneficial information in air quality management in Malaysia.

Materials and Methods

Study Area

This study was conducted at the Pasir Gudang 2 Secondary School (1°28.245'N, 103°53.73'E). Pasir Gudang is an industrial area that is located in the state of Johor in the southern region of Peninsular Malaysia (Figure 1).

Data Collection

12 hourly data of nine chemical parameters and three meteorological parameters over the span of one year (January 2010-December 2010) were analyzed. The data obtained from the Malaysian Department of Environment (DOE) were used to investigate the variability of PM_{10} in the study area. The parameters used in this study were related to meteorological parameters, such as wind speed (WS), temperature (T) and relative humidity (RH). The other contaminants that contribute to air pollution are nitrogen dioxide (NO₂), oxides of nitrogen (NO_x), nitrogen oxide (NO), carbon monoxide (CO), sulfur dioxide (SO₂), non-methane hydrocarbon (NHMC), methane (CH₄) and ozone (O₃). The

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Figure 1: Study area

measurements for all parameters follow the standards set in the Malaysian Ambient Air Quality Guidelines (MAAQG) and NAAQS, such as WS (ms⁻¹), RH (%), T (°C), NO₂ (ppm), SO₂ (ppm), CO (ppm) and PM₁₀ (μ g/m³). PM₁₀ was measured continuously with the Met One Beta Attenuation Monitor (BAM 1020; Washington, USA), licensed by the United States Environmental Protection Agency. The instrument has high resolutions of 0.1 μ g/m³ at 16.7 L/min. The measurement contains the missing value (20.48%) and it was deleted as the missing data is not more than 30% (Liu *et al.*, 2019; Tran *et al.*, 2019).

Data Analysis

Trend analysis is one of the statistical techniques used to determine the variability of pollutants by analysing current trends. In other words, it is a method used to envisage future behaviour by examining the past and the current ones (Manikandan, 2011). The analysis is used as a tool to investigate the changes in pollutant concentration with time (Antilia *et al.*, 2010; Guerreiro *et al.*, 2014; Lang *et al.*, 2019). The statistics used to summarise the characteristics and distribution of the set values are determined by descriptive statistics. There are three types of statistical measures, which are central tendency, the measure of dispersion, and the measure of symmetry (Fernandez et al., 2019). Central tendency refers to the mean, median, and mode (Manikandan, 2011). Median is used to determine the middle values in an order list and also divides the regularity distribution exactly into two parts, which are the 50th percentiles (Manikandan, 2011). The mode is the most frequent score in a data set and the mean, which is used with both discrete and continuous data, is equal to the sum of all the values in the data set divided by the number of values in the data set (Peleg, 2010). A frequency distribution can be measured by measuring the dispersion, which can measure variability, scatter, and the spread of the data collected (Manikandan, 2011). Measures of dispersion are needed when two distributions show the same units and their mean are equal or almost equal, but the variability of the data can be determined through the measurement of the standard deviation (Franzese & Luliano, 2019).

Regression was used in this study as a statistical method, and to investigate the relationship between dependent and independent variable and the independent variable is usually called the regressor variable or predictor variable (Montogometry *et al.*, 2014). This analysis was used to determine the influence of air pollutants on PM_{10} . It is most commonly used in environment particulate exposure evaluation (Montogometry *et al.*, 2014; Latif *et al.*, 2018).

The mathematical model of simple linear regression can be described as in Equation (1).

$$Y = \beta_0 + \beta_1 x + \varepsilon \tag{1}$$

where is the intercept and the slope is, which are unknown regression coefficients, while is the random error with a mean of zero and unknown variance of σ^2 .

PCA is one of the measurable methods that use asymmetrical change to adjust the arrangement of perceptron that potentially correspond to the variables. The arrangements for uncorrelated factors are known as principal components (Samsuddin *et al.*, 2018). PCA can be used as a dimension reduction and data analysis method because of its ability in pattern artificial intelligence, recognition, and data mining (Hoshiai, 1997; Pawlak & Jaroslawki, 2019). The PCA can be defined as in Equation (2).

$$PC_{i} = l_{1i}X_{1} + l_{2i}X_{2} + \dots + l_{ni}X_{n}$$
(2)

where is the principle component and is the loading of the observed variable.

Results and Discussion

The World Health Organisation (WHO) and NAAQS recommend in their air quality guidelines that PM_{10} must be lower than 45 µg/ m³ annually, while the European Commission set a lower value of annual emission at 40 µg/m³. In 2005, the WHO stated that to achieve better air quality, a stricter recommendation for the annual PM₁₀ emission at 20 µg/m³ was proposed. The overall average emission over a 24-hour period for PM_{10} was 46.58 µg/m³ (Table 1). Traffic-related sources are a major contributor to particulate matter, especially in major cities and urban environments. Certain traffic-related particles can be distinguished into exhaust traffic and significantly related with emission of particulate matter with diameter of between 10 μ g/m³ and 510 μ g/m³ (Yang *et al.*, 2019). Winkler et al. (2018) stated that the criteria pollutant emissions generated from fuel combustion

include nitrogen oxides (NO and NO₂, together called NO₂), hydrocarbons (HC), also known as volatile organic compounds (VOCs) or non-methane hydrocarbons (NMHC), carbon monoxide (CO), and particulate matter size smaller than 10 microns, down to 2.5 microns $(PM_{10}-PM_{25})$. The emissions are mostly caused by lubricant volatilisation and incomplete fuel combustion during the combustion procedure and non-exhaust sources, which are generated from non-exhaust traffic movements (Guerreiro et al., 2014; Cao et al., 2019). The non-exhaust emissions are mainly triggered by tires, brakes, road surface wear, or clutches. They exist in the environment as deposited material and become resuspended due to traffic-induced turbulence (Oliviera et al., 2019; Wu et al., 2019). It is estimated that non-exhaust and exhaust sources contribute almost similarly to total traffic-related PM₁₀ emissions and the impact of the number of vehicles on PM is one of the dominant factors (Vardoulakis et al., 2015; Olieviera et al., 2019). The maximum daily average concentration for PM₁₀ in 2010 was 46.58 μ g/m³, while the minimum daily average was 10 µg/m³. The highest daily mean was recorded at 9.00 am. (LT) with the value 63.644 μ g/m³ (28.0-176 μ g/ m³), while the lowest was at 11.00 pm. (LT) at 31.44 μ g/m³ (12-60 μ g/m³) (Figure 2). This is due to the emission of particulate from motor vehicles, industries and dust being release in the study area (Ma et al., 2019) which is located at an industrial area. There are a few studies that stated that the concentration of pollutants are related to space-time random effects based on the situation of the study area (Abdullah et al., 2016; Choi et al., 2018; Chen et al., 2018). Based on Table 1, the maximum value of PM_{10} was caused by haze, biomass burning, and industrial and vehicle emissions (Rani et al., 2018). Malaysia was hit with haze due to forest fires in Indonesia, which also affected the area of study of Pasir Gudang, causing the concentration of PM_{10} to be twice than that of normal conditions, and the event is the main factor behind the maximum value of PM₁₀ (Hamid *et al.*, 2018).

	Valid N	Mean ± SD	Median	Min	Max
CO (ppb _v)	1702	0.509±0.325	0.45	0	3.28
$O_3 (ppb_v)$	1702	0.016±0.012	0.015	0.000	0.11
PM ₁₀ (μgm ⁻³)	1702	46.576±20.990	43.00	10	235
$SO_2 (ppb_v)$	1702	0.0019 ± 0.0021	.00100	0	.043
$NO_{x}(ppb_{y})$	1702	0.013±0.015	.00800	.001	.184
NO (ppb _v)	1702	0.004 ± 0.0110	.00100	0.000	.160
$NO_2(ppb_v)$	1702	0.008±0.006	.00700	0.001	.049
$CH_4(ppb_v)$	1702	1.907±0.273	1.8400	1.50	4.53
NHMC (ppb _v)	1702	0.228±0.147	.1900	.04	1.57
WS (kmhr ⁻¹)	1702	7.306±3.556	6.050	1.0	20.0
T (°C)	1702	27.56±3.1307	26.700	21.7	36.9
RH (%)	1702	80.172±12.075	84.000	46.0	97.0

Table 1: Summary statistics of air pollutants and meteorological parameters







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Figure 2: (a) Time-series plots of CO and median CO, (b) Time-series plots of PM_{10} and median, (c) Timeseries plots of NO₂ and median, (d) Time-series plots of NO and median, (e) Time-series plots of NO_x and median, (f) Time-series plots of SO₂ and median, (g) Time-series plots of NHMC and median, and (h) Timeseries plots of temperature, wind speed and RH

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CO concentration was higher during peak hours in the morning, which is around 6.00 am. (LT) to 10.00 am. (LT), which might be due to the high vehicle volume (Figure 2a). CO is produced from the incomplete combustion of fuels and the main contributor to this pollutant is motor vehicles (Bertrand et al., 2020). Exposure to high CO concentration leads to headaches and increased the risk of adverse cardiopulmonary events, including death (Requia et al., 2018). The trend of gaseous pollutants as shown in Figure 2(c-e) and 2 g shows an increas in the morning, which might come from fuel combustion as the increasing trend of each pollutant occur at the same time as CO, which is around 6.00 am to 9.00 am. (LT). The development of vehicular emission inventory is necessary to assess the emissions in this area (Pinto et al., 2020). Figure 2(f) shows the trend of sulphur dioxide (SO₂), which is suspected to have come from industrial activities as this pollutant is generally the result of anthropogenic activities (Fang et al., 2018). The main source emission of SO_2 is the combustion of sulphur compounds, combustion of fossil fuels (mainly coal and heavy oils), biomass burning, and the smelting of sulphur containing ores (Oh et al., 2019). The increase of SO, in the morning is due to operations starting, which usually occurs in the early operating hours and evening during shifting time.

The average temperature was 27.56°C and it ranges between 21.7°C and 36.9°C (Table 1). The minimum and maximum RH are 46% and 97% for Asian countries (WHO, 2018). The average wind speed is 6.05 km/h. The diurnal variation of T, RH, and WS are shown in Figure 2(h). The temperature and RH show an opposite diurnal pattern. The RH shows a downward trend from 8 a.m. to 2.00 p.m. local time, while the temperature starts to increase at that time. The trends for both meteorological factors show that the air in the study area was dry and warm in the afternoon. The ambient temperature was at 25.5°C as RH reaches the highest level, which is 90% at midnight. Figure 2(h) also shows that there is a drastic increase in temperature around 10.00 am and a decreasing percentage of RH in the afternoon. The trend of PM_{10} , as shown in Figure 2(b), matches the ozone precursor concentrations, as seen in Figures 2(c), 2(d), 2(e) and 2(g) as it indirectly affects aerosol concentrations. CH₄ and NHMC exhibit stable emissions by the chemical formation of secondary organic aerosol and both of these contaminants indirectly influence the variability of aerosol concentration, which is PM_{10} (Chen *et al.*, 2018; Oh *et al.*, 2019; Stamatelopoulou *et al.*, 2019).

Figure 3 shows the simple linear regression of all variables with PM₁₀, in which all the parameters are statistically significant. There are significant positive correlations between PM₁₀ and SO₂ (R²=0.0302, p<0.01), CH₄ (R²=0.0817, p<0.01), RH (R²= 0.0581, p<0.01), NO_v (R²=0.1118, p<0.01) and NO (R²=0.0786, p<0.01). A simple linear regression model linking PM₁₀ with the other variables was used and it is a feasible approach to evaluate the particulate emission in the study area (Vardoulakis et al., 2015). These regression analyses show that the increase in pollutants in the study area increase the PM₁₀ concentration and proved that nonexhaust and exhaust sources contribute almost correspondingly to the total traffic-related PM₁₀ emissions. There is a negative relationship between PM₁₀ and temperature as the weight of PM₁₀ during low temperature is heavier and, hence, it is suspended on the ground (Yang et al., 2019; Yue et al., 2019). Most countries in Asia found a negative correlation between PM₁₀ concentration and wind speed, RH and temperature (Yue et al., 2006; Das et al., 2018).

PCA requires Kaiser Meyer Olkin (KMO) of Sampling Adequacy to be more than 0.50 for the arrangement factors and the Bartlett's test of Sphericity probability must be <0.05 (Azimi *et al.*, 2018). Table 2 shows the KMO and Bartlett's test values, which satisfy the requirements for PCA.

Table 3 lists the eigenvalues related to each linear component following extraction and rotation. The eigenvalue related to each factor addresses the distinction cleared up by particular linear components and demonstrates their eigenvalue in terms of percentage of variance.



Figure 3: (a) Correlation between PM_{10} and SO_2 , (b) Correlation between PM_{10} and RH, (c) Correlation between PM_{10} and NO_2 , (e) Correlation between PM_{10} and NO, and (f) Correlation between PM_{10} and temperature

Kaiser-Meyer-Olkin Measure o	.752	
Bartlett's Test of Sphericity	Approx. Chi-Square	31690.887
	df	66
	Sig.	.000

Utilising 3 factors, the variability of the data set was 75.487%. The rotation has the effect of enhancing the factor structure and one outcome for this data has relative hugeness of the three components. Before rotation, factor 1 (48.650%) is significantly more different than factor 2 (18.420%) and factor 3 (8.417%). However, after extraction, it accounts for only factor 1 (35.505%), factor 2 (28.975%) and factor 3

(11.09%). The rotated matrix rotation utilising varimax with Kaizer normalization is shown in Table 4. The loading factor of more than 0.5 is considered strong, 0.4 to 0.49 moderate and less than 0.3 weak. In general, PC-1 has a strong contribution to traffic-originated emission of PM_{10} , CO, NO₂, NO_x, and NHMC. The results from Table 4 show that the site contains PM_{10} , CO, and NO₂, which are mainly caused by

local traffic emissions (Mohamed *et al.*, 2016). Urban and industrial areas have significant contamination, which for the most part is contributed by engine vehicles (Azimi *et al.*, 2018). Thus, it indicates that PC-1 is associated with local traffic sources, with a contribution of 35.505%.

PC-2 is composed of the parameters of O_3 , wind speed, RH and temperature. All meteorological parameters are based on PC-2. The contribution of ambient temperature and wind speed in the industrial area has a strong negative relationship with RH. Meteorological factors are connected to explaining the pollutant variability in Malaysia. The contribution for

Component	onent Initial Eigenvalues		Extraction Sums of Squared		Rotation Sums of Squared Loadings				
	Total	% of Variance	Cumulative (%)	Total	% of Variance	Cumulative (%)	Total	% of Variance	Cumulative (%)
1	5.838	48.650	48.650	5.838	48.650	48.650	4.261	35.505	35.505
2	2.210	18.420	67.070	2.210	18.420	67.070	3.477	28.972	64.478
3	1.010	8.417	75.487	1.010	8.417	75.487	1.321	11.009	75.487
4	.874	7.286	82.773						
5	.751	6.255	89.028						
6	.383	3.189	92.217						
7	.361	3.008	95.225						
8	.243	2.027	97.251						
9	.175	1.457	98.708						
10	.113	.945	99.653						
11	.042	.347	100.000						
12	3.244E-005	.000	100.000						

Table 3: Total variance explained

	Component				
	1	2	3		
СО	-	-	-		
O_3	-	.787	-		
PM_{10}	-	-	-		
SO_2	-	-	.929		
NO _x	.924	-	-		
NO	.928	-	-		
NO ₂	.583	-	.513		
CH_4	.586	-	-		
NHMC	.859	-	-		
WS	-	.838	-		
Т	-	.929	-		
RH	-	944	-		

Table 4: Rotated component matrix

PC-2 was 28.972%. PC-3 consists of high concentrations of SO₂ and NO₂ This is possible because the area is located in the industrial area and many studies have found that emissions of SO, are caused by combustions due to the uneven evolution of industrial activities (Abdullah et al., 2016; Ma et al., 2019). The acidification caused by the emission of SO₂ can cause damage towards crops and plants and indirectly disturbs the quality of soil and water, besides the air in an area that is exposed to high emission of SO, (Yue et al., 2006; Azimi et al., 2018; Cao et al., 2019) through which their harmful effects result in serious human health. SO, and NO, were precursors of a PM that come from different areas and poses significant challenges in the future (Azimi et al., 2018; Franzese & Luliano, 2019; Kalimeri et al., 2019). PC-3 contributes 11.009% in the study area, which are caused by industrial activities.

Conclusion

This study investigates the trend, interrelation, and source apportionment of PM_{10} concentrations with its influencing factors; trace gases and meteorological parameters. The diurnal trend of PM₁₀ concentrations in this industrial area had the highest peak during the morning. Other trace gases also had the same diurnal trend, which may be due to emissions from the combustion of motor vehicles. Simple linear regressions proved that several trace gases and meteorological parameters had a significant influence on the PM₁₀ concentration. The air quality in this industrial area was influenced by traffic emissions (35.5%), weather (29.0%)and industrial activities (11.0%). In conclusion, the results can be used for air quality abatement measures in combating air pollutant sources at the root cause.

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