The cause-and-effect analysis of ground level ozone (O_3), air pollutants and meteorological parameters using the causal relationship approach

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ABSTRACT

The recent reduction in ambient air quality has been related to anthropogenic activity. This anthropogenic activity played a big role in polluting the atmosphere. Ozone (O_3) is one of the air pollutants that is not emitted directly from the source at ground level. Anthropogenic activities like industrial and mobile sources, on the other hand, may produce O_3 pollutant precursors directly. Human health, the environment, materials, and crops have all been found to be negatively affected by this O_3 pollutant. As a result, this study investigated the causal relationship between O_3 and particulate matter, gaseous pollutants, and meteorological circumstances. Three monitoring stations, each representing a different geographic region, were chosen. The three sampling monitoring stations are in Negeri Sembilan, Kelantan, and Perlis, and represent industrial, urban, and sub-urban areas, respectively. Sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and wind speed (WS) are all causally related to O_3 in Nilai, as are SO_2 and carbon monoxide (CO) in Kota Bharu, and NO_2 and CO in Kangar. In the meantime, it was discovered that the causal relationship between the causative parameter and

O_3 is only one-way. The O_3 considers this to be a secondary contaminant that may require this parameter in order to form in ambient air. However, none of the primary parameters have a directional relationship to the others, except for O_3. As a result, this discovery may be useful in future research to increase our understanding of air quality, particularly the O_3 pollutant.

Keywords: Air pollutant; causality relationship; ground level ozone; $O_{3;}$ Malaysia

INTRODUCTION

The rapid economic development brought about by the acceleration of industrialisation and urbanisation has resulted in an increase in air pollution as a result of pollutant emissions (Ning et al., 2018). According to the World Health Organization (2016), high-income countries are the most affected by increasing industry and urbanisation when compared to middle- and low-income countries. Unfortunately, compared to America, Europe, Africa, and the Caribbean, the magnitude of urbanisation in Asia was unparalleled (Roth et al., 2011). China has been identified as the country with the fastest-growing urbanisation in terms of population (Chen et al., 2016).

A recent study in China discovered that, over the course of a decade, ambient air quality across the country has deteriorated due to an increase in industrial activities (He et al., 2019; Zhu et al., 2019). The vastly increasing numbers of industrial factories to provide and accommodate human demands, not just in China, but globally. As a result, pollutant emissions from industrial activities were found to be linked to air pollution levels (Sun et al., 2020; Al-Joboori et al., 2020). In the ambient air, there are three major sources of air pollutants: stationary sources, mobile sources, and natural sources (Raffee et al., 2018). Stationary sources include industrial activities and power plants. Meanwhile, a mobile source is defined as emissions from a vehicle, aircraft, ships, or any method of transportation that uses combustion fuel, whilst a natural source is defined as forest fires and volcanoes, which are the most common causes of haze.

Air pollution is closely related to decreases in ambient air quality. The word "air pollution" refers to the presence of air pollutants in the ambient air at levels that impose to health hazard (Hassoun et al., 2019). The pollutant can be a gas, a liquid, or a particle that disperses in the environment. The air pollution study has been inspired for a decade by the dangers of air pollution to human health. Whereas the findings show that well-known air pollution has a negative impact on human health and welfare (Kampa & Castanas, 2008).

In China, recent air pollution research has shifted its focus to ground-level ozone (O_3), which has surpassed particulate matter as the most prevalent air pollutant as a result of anthropogenic activities such as industrial and urbanisation processes (Lu et al., 2019). In addition, the O_3 pollutant is of particular concern because it poses a greater risk to human health than other air pollutants. Based on its features, this O_3 was classed as a secondary pollutant rather than a primary pollutant.

The main feature of O_3 pollutant forms when its precursors, namely volatile organic compounds (VOC) and nitrogen oxides (NO_X), react in the presence of solar radiation (sunlight). This oxidation process was led to form a dangerous gaseous of O_3. Thus, people living in areas that repeatedly recorded exceeding permissible O_3 limit were more exposed a greater risk health effect. Whereas, according to Pierre et al. (2017), the O_3 pollutant in Europe recently considered as one of the most dangerous air pollutants and it can be worse in the future. This is due to O_3 pollutant was not only affect to human health, despite the environment and materials.

Jerrett et al. (2009) reported on the relationship between O_3 concentration and the long-term health effects of O_3 exposure in humans. The discovery was widely used in most studies on O_3 and human health. In 2014, the total number of premature deaths recorded in China was 89,391 due to the respiratory condition Chronic Obstructive Pulmonary Disease (OCPD) caused by O_3 exposure (Lin et al., 2018). Studies on O_3 pollutant have been increasingly relevant in recent years as a result of concerns about the harmful effect toward human health. Because O_3 does not exist alone in ambient air, numerous air quality studies have focused on the relationship between this pollutant and other factors such as PM_{10}, sulphur dioxide (SO_2), nitrogen dioxide (NO_2), carbon monoxide (CO), wind speed (WS), relative humidity (RH), and temperature (T) (Zhang et al., 2017; Raffee et al., 2018; Awang et al., 2018).

The most frequent method for examining correlations between O_3 pollutants and other variables is correlation analysis. Few works of literature had been published regarding the study, such as the finding of a strong positive association in three separate locations in an urban region, indicating that temperature influences O_3 pollutant concentrations (Awang et al., 2018). Hu et al. (2019) discovered that the O_3 pollutant was substantially positively associated to particulate matter and CO in a sub-urban area.

Furthermore, the correlation analysis revealed strong relationships between air pollutants such as O_3, PM_{10}, CO, and NO_2 and meteorological parameters (such as wind speed, temperature, and relative humidity) in an urban setting (Rahman et al., 2015). According to the research, O_3 may have correlations with other parameters, although at a different level of association. These findings demonstrate the need of looking into the relationships between other parameters and O_3 dispersion in air quality studies.

The correlation analysis methodology was commonly used in air quality applications to describe the relationship of O_3 pollutant to other factors such as NO_2, CO, NO_X, and meteorological data. As a result, it looked at the correlations between each factor and the level of O_3 concentration. Moreover, this statistical technique, on the other hand, merely reveals the level of correlations between dependent parameters and the parameters that were considered. Furthermore, a correlation analysis of two variables is insufficient to determine the links between more than two variables. On the other hand, there may be an existence that is unable to account for the third and subsequent factors (Granger, 1969).

Thus, causal relationship analysis is a statistical technique that can help to solve such a problem. This method implies a dependency relationship between the cause and effect of each parameter considered. Simultaneously, the statistical correlation technique revealed only the relationship between the parameters. By comparing the statistical techniques of correlation and causal relationship, the causal relationship examines the directional cause and effect of each parameter and provides the significant value of directional to each other. Rather than simply reporting the relationship between parameters, the causal relationships statistical technique and the directional of each parameter can be explained in terms of the level cause and effect to dependent parameter. Due to this issue, the current study used the causal relationship to investigate the significant directional as well as the cause and effect of O_3 concentration with particulate matter pollutant, gaseous pollutant, and meteorological parameters at three different locations in Malaysia, including an industrial, urban, and suburban area. The findings and results may be useful for other researchers and the government in providing early mitigation for future studies due to the possibility of increasing anthropogenic activities that may be the cause of increasing O_3 pollutants' levels.

AIR QUALITY DATA

Secondary data on air quality was obtained from Malaysia's Department of Environment (DoE). This data was continuously recorded and controlled from the automated air quality control remote station while going through the process for established standards required by the Malaysian Department of Environment. From January 2006 to December 2017, hourly averages of ground level ozone (O_3), meteorological parameters (wind speed, temperature, and relative humidity), particulate matter (PM_{10}), and other gaseous pollutants (CO, NO_2, SO_2) were obtained and converted to monthly average data.

Numerous studies have attempted to explain fluctuations in O_3 concentrations as a result of gaseous pollutants and volatile organic compound (VOC) precursors, therefore the historical data obtained from the Department of Environment was useful (Ismail et al., 2016; De Souza et al., 2017; Apondo et al., 2018). CO, SO_2, and NO_2 were created as major gaseous pollutants in the atmosphere before being chemically oxidised to O_3 in the presence of solar light. O_3 and its precursors were transported and accumulated by the wind (Teinilä et al., 2019). The high wind speed may reduce the O_3 concentration, allowing the pollutant to travel to new locations.

LOCATION OF STUDY AREA

Three Malaysian air monitoring stations were chosen for this study. There are various types of locations and regions in Peninsular Malaysia. The first air quality monitoring station located in Nilai (N02º 15.924', E102º 10.554') state of Negeri Sembilan. The Malaysian Department of Environment classified this air quality monitoring station as industrial. Nilai air quality monitoring stations are strategically located in rapidly expanding industrial areas, resulting in significant air pollution (Ahmat et al., 2015). The second air quality monitoring station classified as urban type located in Kota Bharu (N06º 09.520' E102º 15.059') in state of Kelantan. The Kota Bharu air quality monitoring station is in northeastern part of peninsular Malaysia, close to Thailand border. Kota Bharu's main activities are trading and tourism (Masseran et al., 2016). The last air quality monitoring station selected in this study is Kangar (N06° 19.545' E99° 51.311'). Kangar is the capital of Perlis and located at southern part of Peninsular Malaysia. There are sub-urban area with numerous human activities and undergoing rapid urban development (Abdullah et al., 2017). The selected air quality monitoring stations in this current study has been notable the station as S1, S2 and S3 for Nilai, Kota Bharu and Kangar respectively. The geographical map details for all three monitoring stations showed in Figure 1.

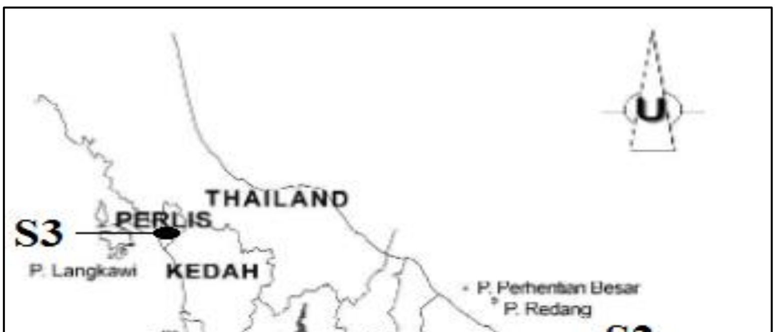

Figure 1. Geographical map for all three selected sampling stations

METHOD OF ANALYSIS

The causal relationship statistical technique maximises the relationship between cause and effect between dependent and independent parameters taken into consideration in this study. This could provide an important result of the cause and effect of each independent parameter on the dependent parameter, which is reported in terms of significance and direction. The record data was required to check the stationary of each of the data in order to comply with the causal analysis. The monthly records data worth of 144-months were utilized. Data of O_3, PM_{10}, gaseous pollutants and meteorological parameters was subjected to the stationary test and there are numerous statistical tests that can be used to determine whether variables in a multivariate time series are stationary.

The most commonly used time series purposed test is the Augmented Dicky-Fuller (ADF) test (Abdel-aziz & Frey, 2003). The ADF test is represented by the following equation:

$$\text{ADF} = \alpha_0 + p_1 y_{t-1} + \sum_{j=2}^{p-1} \beta_j \nabla y_{t-j} + e_t \tag{1}$$

where,

α_0 : Drift Component

e_t : Independent and homogeneous error term

In order to determine the stationary of the series, Sansuddin et al. (2011) stated that the hypothesis as follows:

H_o : the time series data is non-stationary

H_1 : the time series data is stationary

where, the H_o can be rejected by if the significance value (p) is smaller or equal to 0.05.

The causality statistical test can then be performed after the stationary test has been applied to all data. The following equation is used to estimate the causality statistical test (Rahmah & Kashem, 2017).

$$y_t = g_0 + a_1 y_{t-1} + ... + a_p y_{t-1} + b_1 x_{t-1} + ... + b_p x_{t-p} + u_t \tag{2}$$

$$x_t = H_o + c_1 x_{t-1} + ... + c_p x_{t-p} + d_1 y_{t-1} + ... + d_p y_{t-p} + v_t \tag{3}$$

Then, testing H_0: $b_1 = b_2 = ….. = b_p = 0$, against H_A: x Granger causes y. Similarly, testing H_0: $d_1 = d_2 = ….. = d_p = 0$, against H_A: y_t Granger causes x_t. The H_0:b_1 represent the dependent series while the H_o:d_1 represent the independent series. While a is the coefficient values of the series. In each case, a rejection of the null implies there is Granger causality. In other way, the Granger causality can be determine using the statistical F-statistics and the hypothesis for Granger causality test as follows (Jordaan & Eita, 2009):

H_o : the series is not granger-cause

H_1 : the series is granger-cause

If the significance F-statistics value is equal to or less than 0.05, the null hypothesis is rejected, indicating that the dependent series was granger caused by the independent series.

The results of a causal statistical test, on the other hand, can be represented graphically. This will necessitate a thorough examination of the causes and effects of each parameter in the study. The graphical representation of a causal statistical test between parameters can be unidirectional or bidirectional. Because the significant test for 0.05 was used in this study, the unidirectional indicate that the independent parameter was not a cause and effect to the dependent parameter or vice versa.

Furthermore, if the parameter did not have a Granger cause to O_3 at a significant level of 0.05, it may be significant at a level greater than 0.05. Meanwhile, the bidirectional significant the dependent and independent parameter had a relationship that caused and effect to each other at 0.05 significant test. As a result, the directional was affected by the significant value. Figure 2 depicts the details of unidirectional and bidirectional interactions. According to the diagram, the dot direction lines indicate that the parameter B had a cause-and-effect relationship with the direction of the line to the parameter A. At the same time, the straight direction line indicates that the parameter A did not have a cause-and-effect relationship with the direction of the line to the parameter B.

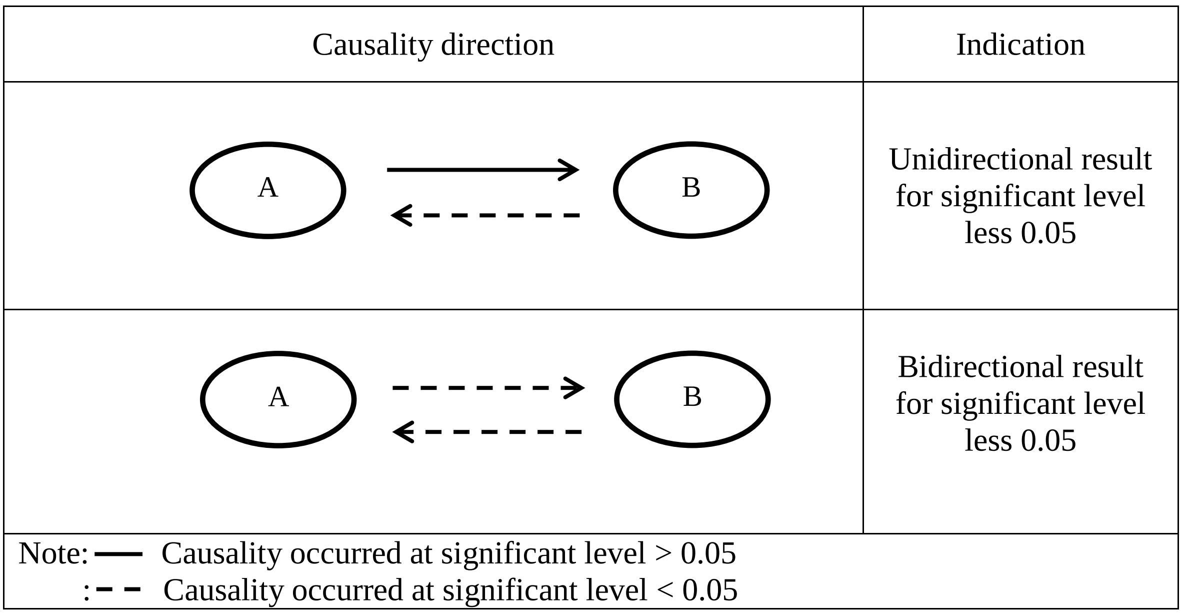

Figure 2. The direction of illustration of causal relationship statistical test

RESULTS AND DISCUSSION

The descriptive statistics of O_3 concentration data from January 2006 to December 2017 were presented in Table 1. The standard deviation for all three sampling monitoring stations was recorded in ranges ranging from 0.0155 ppm to 0.0192 ppm, indicating that the concentration variability of O_3 concentration was recorded almost identically. Meanwhile, the mean values in all three sampling monitoring stations are greater than the median, and the data is skewed to the right, indicating that moderate O_3 concentrations were recorded.

The maximum O_3 concentration recorded at Nilai, Kota Bharu, and Kangar monitoring stations is 0.1140 ppm, 0.0830 ppm, and 0.0810 ppm, respectively. Thus, the data of O_3 concentrations at industrial sampling stations were found to be higher than the Malaysia Ambient Air Quality Guideline (MAAQG) when compared to urban and sub-urban sampling stations. The high recorded concentrations of O_3 in industrial sampling stations are not surprising due to industrial emissions of NO_X and volatile organic compound (VOC) as precursors of O_3, which are primarily emitted from industrial processing and heavy transportation activities (Hidy et al., 2015). Furthermore, the disparities in concentrations observed across all three sampling monitoring stations could be attributed to differences in local emission from anthropogenic activities of mobile and stationary sources in atmospheric compositions (Banan et al., 2013).

Table 1. The descriptive statistics results of O_3 for all three sampling monitoring stations

Station	Standard Deviation	Mean	Median	Skewness	Maximum
Nilai (S1)	0.0155	0.0157	0.0100	1.6070	0.1140
Kota Bharu (S2)	0.0145	0.0170	0.0100	0.8010	0.0830
Kangar (S3)	0.0127	0.0210	0.0190	0.6620	0.0810

The hourly average data was used to analyse the O_3 concentration behaviour pattern in depth. The diurnal dispersion of O_3 concentration was depicted in Figure 3 for all three sampling monitoring stations. The O_3 concentration in Kota Bharu and Kangar sampling monitoring stations began to rise at 9.00 a.m., peaked at 2.00 p.m., and began to fall at 4.00 p.m. Meanwhile, the Nilai sampling monitoring station recorded a peak one hour later than the Kota Bharu and Kangar sampling monitoring stations, which recorded at 3.00 pm and slightly decreased time to time started at 4.00 pm.

According to the diurnal graph results, the industrial area has a higher recorded O_3 concentration than the urban and suburban areas. The concentration of precursor at the industrial sampling station was thought to be the primary cause of the disparities in sampling monitoring station results. Because of the intensity of solar radiation (sunlight) in Malaysia, the peak of O_3 concentrations occurred during the day between 1.00 pm and 3.00 pm (Abdullah et al., 2019; Awang et al., 2018). The findings were also confirmed by Geng et al. (2008), who determined that the high intensity of solar radiation was the primary contributor to the high recorded O_3 concentration values.

As a result of the different diurnal dispersion of O_3 observed at all three sampling stations, a further analysis using the causal relationship statistical technique was performed to determine the parameter that might influence the level of O_3 concentration. Typically, causal relationship analysis is based on the stationary of the data from the records. Mills (2015) defines stationary data as the mean and variance of a data set that does not change over time.

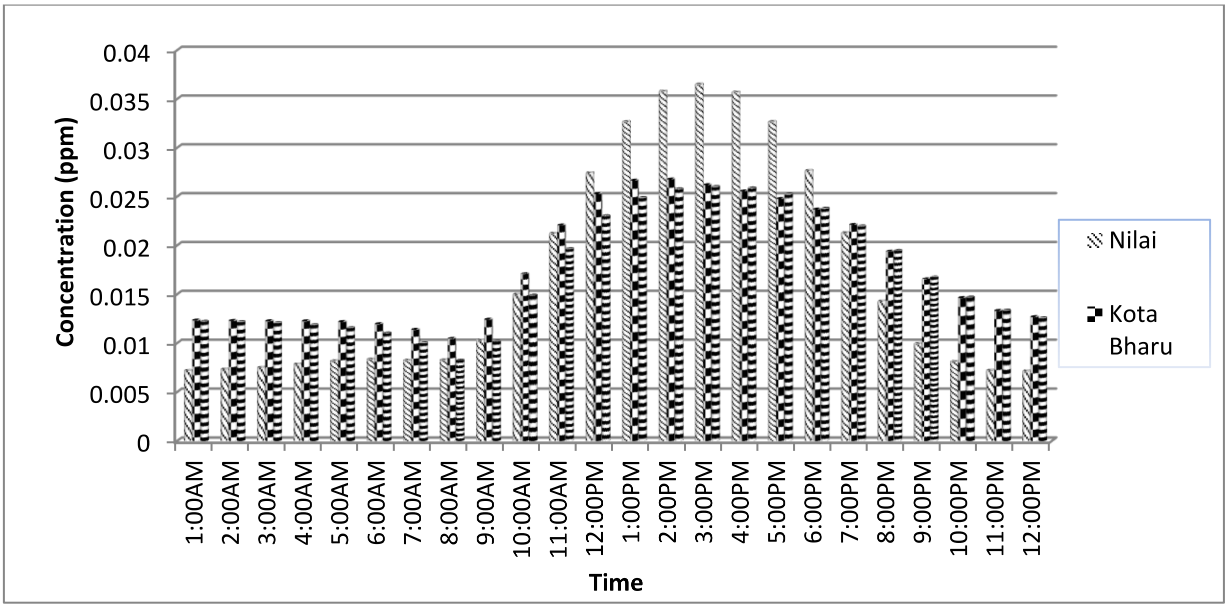

Figure 3. Diurnal dispersion of O_3 concentration at all three sampling monitoring stations

Physical observations using the series plot graph may not always be able to provide clear information about the stationary of each set of record data. To be more specific, the statistical Augmented Dicky-Fuller (ADF) test was run on all data sets in all sampling monitoring stations to examine the stationery. The ADF test was adequate for using and checking the stationary of the current data; either it is stronger to reject the hypothesis at a given level of confidence (Omar et al., 2013).

Table 2 shows the t-statistic and p-value result of ADF test for non-stationary results for record data of air pollutants and meteorological data sets. According to the ADF test, all parameters at the Nilai sampling monitoring station had non-stationary data records. Furthermore, Kota Bharu and Kangar's remaining sampling monitoring stations had six and five stationary parameters, respectively. Temperature and relative humidity are the non-stationary parameters in Kota Bharu, according to this. Meanwhile, at Kangar, the parameters of ground-level ozone, nitrogen dioxide, and relative humidity were recorded as non-stationary data. As a result of the non-stationary obtained for several parameters, differencing is required to convert the data to a stationary data set in order to comply with the rules in causality methodologies, which require that the data be stationary.

The results of the causality relationship are shown in Table 3. According to the findings, three parameters, specifically SO_2, NO_2, and WS, show a causative relationship with O_3 concentration in Nilai at a significant level less than 0.05, with p-values of 0.0132, 0.0086, and 0.0475, respectively. Meanwhile, both sampling stations in Kota Bharu and Kangar measured two causative parameters to O_3, namely SO_2 and CO in Kota Bharu and NO_2 and CO in Kangar, at a significant level of 0.05.

Table 2. The results of ADF statistic and *p*-value for non-stationary data set for all three sampling monitoring stations

Station	Parameter	t-statistics	*p*-value

Nilai monitoring station	O_3	-0.9403	0.0713
	SO_2	-1.1727	0.0157
	NO_2	-0.3554	0.8880
	CO	-0.5372	0.0727
	PM_{10}	-1.1503	0.0490
	WS	-1.8985	0.1636
	T	0.9449	0.1546
	RH	-0.9389	0.2589
Kota Bharu monitoring station	T	-0.4819	0.6795
	RH	-0.5151	0.1079
Kangar monitoring station	O_3	-1.4860	0.1280
	NO_2	-1.2538	0.2068
	RH	-0.3135	0.6065

The details cause and effect between parameters in directional causality, on the other hand, were also obtained. The results of the directional causality test are the major finding that distinguishes this study from previous literature on correlation studies in air quality. Figure 4 depicts the outcome of the parameters directional. The causality test was found to be significant at 0.05 because no bi-directional events occurred. At a significance level of less than 0.05, the presented figure shows that in the Nilai sampling monitoring station, one causality directional was found to O_3 pollutant from NO_2, SO_2, and WS. Furthermore, SO_2 and CO were found to be the causal parameters for O_3 in Kota Bharu, while at Kangar, NO_2 and CO were found to be the causal parameters.

According to Figure 4, three parameters have cause and effect at a 0.50 significance level at Nilai and Kota Bharu. Furthermore, the cause-and-effect relationship between SO_2 and NO_2 at Nilai was observed to have a bi-directional effect. At a significance level of 0.05, the causal relationship analysis revealed that air pollutants concentrations and meteorological parameters had varied causation relationships in each air quality monitoring station. However, as it is determined as the precursors of O_3 pollutant concentration, the parameters of gaseous pollutants such as SO_2, NO_2, and CO, are likely to have a causal relationship with O_3.

Thus, the results of the causality relation in the industrial area (Nilai) are expected due to regional and local emission, which refers to transporting emission and motor vehicle emission, respectively, while in the urban area (Kota Bharu) and sub-urban area (Kangar) monitoring stations, the results are expected due to regional and local emission, which refers to the transporting emission and motor vehicle emission, respectively. At the same time, the transfer of emissions by wind from nearby places such as industrial to urban or sub-urban areas causes significant levels of O_3 pollution.

Table 3. The causality relationship t-statistic and *p*-value for all sampling monitoring stations

Nilai Sampling monitoring station							
Parameter	SO_2	NO_2	CO	PM_{10}	WS	T	RH
t-values	-2.515	-2.673	0.696	0.017	1.918	-0.439	-0.850
p-value	**0.0132**	**0.0086**	0.4879	0.9867	**0.0475**	0.6616	0.3971
Kota Bharu Sampling monitoring station							
Parameter	SO_2	NO_2	CO	PM_{10}	WS	T	RH
t-values	2.1550	-0.5040	3.5350	-3.4240	0.9680	-0.1760	-0.2250
p-value	**0.0331**	0.6155	**0.0006**	0.0841	0.3349	0.8604	0.8223
Kangar Sampling monitoring station							
Parameter	SO_2	NO_2	CO	PM_{10}	WS	T	RH
t-values	-1.3510	3.3300	-2.6590	-2.6080	-0.5910	-0.4510	-0.2250
p-value	0.1792	**0.0011**	**0.0089**	0.1102	0.5554	0.6529	0.8223
*Bold indicate as parameter caused that influence O_3 concentration at significant level less than 0.05							

CONCLUSION

The study's findings show that industrial monitoring station had higher O_3 concentrations than urban (Kota Bharu) and sub-urban monitoring stations (Kangar). The findings also revealed that the maximum O_3 concentrations that exceeded the MAAQG acceptable limit were only found in Nilai. Meanwhile, in Nilai, Kota Bharu, and Kangar, the diurnal dispersion of O_3 concentration follows a similar trend. The O_3 concentration, on the other hand, peaks at midday following a quick rise in the morning, then gradually declines to a low level in the evening. The results of a causal association between the O_3 pollutant and other parameters (such as PM_{10}, SO_2, NO_2, CO, T, WS, and RH) indicated that the gaseous pollutants such as

SO_2, NO_2, and CO were causal to the O_3 concentration at a significant level less than 0.05. At Nilai, wind speed was the sole meteorological parameter that was causative to O_3 at a significant level up to 0.05. However, all directions of a causal relationship in all selected monitoring stations were examined in unidirectional only, except for Nilai between SO_2 and NO_2 at a significant level, not more than 0.5. The finding is expected due to the mechanism of the O_3 pollutant, which needs this parameter to build up in ambient air. By defining the parameters that influence O_3 concentration using the causal relationship developed in this study, it is hoped that other researchers will be able to improve air quality studies, particularly for prediction purposes.

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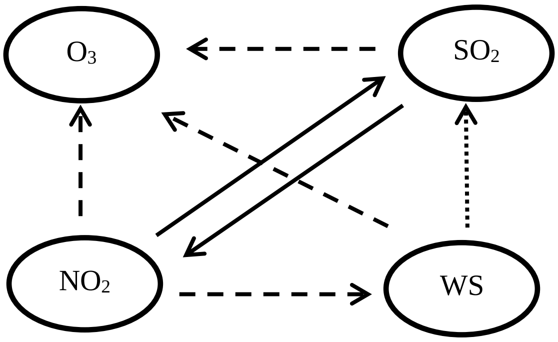
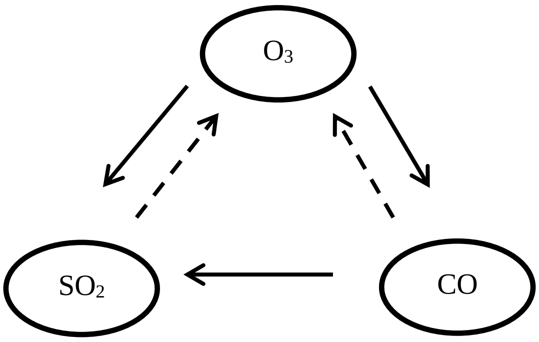
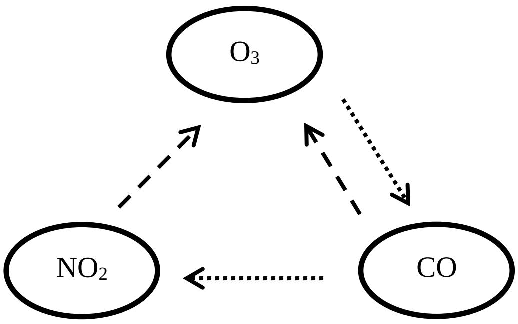
Granger causality direction	Monitoring Station
	Nilai monitoring station
	Kota Bharu monitoring station
	Kangar monitoring station

Note: – – Causality occurred at significant level < 0.05
: ...... Causality occurred at significant level < 0.10
: —— Causality occurred at significant level < 0.50

Figure 4. The direction of causality for all sampling monitoring stations

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