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Cause-and-effect analysis of ground-level ozone, air pollutants, and meteorological parameters using the causal relationship approach

Ahmad Fauzi Raffee*, Hazrul Abdul Hamid*, Siti Nazahiyah Rahmat and Muhammad Ismail Jaffar

*Universiti Tun Hussein Onn, Malaysia

** School of Distance Education, Universiti Sains Malaysia

**Correspondent author: hazrul@usm.my

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ABSTRACT

Recent deterioration of ambient air quality is related to anthropogenic activities, which play a significant role in atmospheric pollution. Ozone (O_3) is an air pollutant that is not emitted directly from sources at the ground level. Meanwhile, anthropogenic activities, such as industrial and mobile sources, may directly produce O_3 pollutant precursors. Human health, the environment, materials, and crops are negatively affected by O_3 pollutants. Therefore, the present study investigated the causal relationships between O_3 and particulate matter, gaseous pollutants, and meteorological conditions. Three monitoring stations, each representing a different geographical region, were selected. The three monitoring stations were in Negeri Sembilan, Kelantan, and Perlis, representing industrial, urban, and suburban areas, respectively. Sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and wind speed (WS) were causally related to O_3 in Nilai; SO_2 and carbon monoxide (CO) in Kota Bharu; and NO_2 and CO in Kangar. However, the causal relationship between the causative parameters and O_3 was one-way. Therefore, O_3 is considered to be a secondary contaminant that may require these parameters to be formed in the ambient air. However, none of the primary parameters showed a directional relationship with the other parameters, except for O_3. These findings may be useful in future research to improve our understanding of air quality, particularly the status of O_3 pollutants.

Keywords: Air pollutant; Causal relationship; Ground-level ozone; O3; Malaysia

INTRODUCTION

Rapid economic development resulting from by the acceleration of industrialization and urbanization has resulted in an increase in air pollution due to pollutant emissions (Ning et al., 2018). According to the World Health Organization (2016), high-income countries are more affected by increasing industrialization and urbanization than middle- and low-income countries. Unfortunately, compared with the Americas, Europe, Africa, and the Caribbean, the magnitude of urbanization in Asia is unparalleled (Roth et al., 2011). China has been identified as the country with the fastest growing urbanization 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 owing to an increase in industrial activities (He et al., 2019; Zhu et al., 2019). The vastly increasing number of industrial factories provide and accommodate human demands not only in China but also around the globe. Pollutant emissions from industrial activities are linked to air pollution levels (Sun et al., 2020; Al-Joboori et al., 2020). The ambient air has three major sources of air pollutants: stationary, mobile, and natural sources (Hamid et al., 2013). Stationary sources include industrial activities and power plants. Meanwhile, mobile sources include emissions from vehicles, aircraft, ships, or any form of transportation that uses combustion fuel. Meanwhile, natural sources include forest fires and volcanoes, which are the most common causes of haze.

Air pollution is closely related to the decrease in ambient air quality. The term "air pollution" refers to the presence of air pollutants in ambient air at levels that impose health hazards (Hassoun et al., 2019). Pollutants can include gases, liquids, or particles dispersed in the environment. Studies have focused on air pollution over the past decade because of its human health hazards, and the negative effects of air pollution on human health and welfare have been documented (Kampa & Castanas, 2008).

In China, the focus of recent air pollution research has shifted to ground-level ozone (O_3), which has surpassed particulate matter as the most prevalent air pollutant due to anthropogenic activities, such as industrial and urbanization processes (Lu et al., 2019). O_3 pollutants are of particular concern because they pose a greater risk to human health than the other air pollutants. Based on these features, O_3 is classified as a secondary rather than primary pollutant.

The primary feature of O_3 pollutants is the formation of volatile organic compounds (VOCs) and nitrogen oxides (NO_X), which react with solar radiation (sunlight). This oxidation process leads to the formation of dangerous gaseous O_3. Thus, people living in areas that repeatedly exceed the permissible O_3 limits are at a greater risk of health adversities. According to Pierre et al. (2017), O_3 pollutants were recently declared one of the most dangerous air pollutants in Europe, and O_3 pollution may worsen in the future. In addition to human health, O_3 pollutants affect the environment and materials.

Jerrett et al. (2009) reported a link between O_3 concentration and long-term health effects of O_3 exposure in humans, and this link has been increasingly discovered in most studies on O_3 and human health. In 2014, the total number of premature deaths from chronic obstructive pulmonary disease (OCPD) caused by O_3 exposure recorded in China was 89,391 (Lin et al., 2018). Studies on O_3 pollutants have become increasingly relevant in recent years because of concerns regarding their harmful effects on human health. As O_3 does not exist alone in ambient air, numerous air quality studies have focused on the association between this pollutant and other factors, such as PM_{10}, sulfur 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).

Correlation analysis is the most frequently used method for examining the association between O_3 pollutants and other variables. However, few studies have been published on this topic. For instance, Awang et al. (2018) noted a strong positive association between temperature and O_3 pollutant concentrations at three separate locations in an urban region. In another study, Hu et al. (2019) discovered that O_3 pollution is positively associated with particulate matter and CO in a suburban area.

Furthermore, a correlation analysis revealed strong associations 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 previous studies, O_3 may be correlated with other parameters, albeit at different degrees of association. These findings demonstrate the need to examine the association between other parameters and O_3 dispersion in air quality studies.

Correlation analysis is commonly used in air quality applications to describe the association between O_3 pollutants and other factors, such as NO_2, CO, NO_X, and meteorological data. Therefore, we can examine the correlation between each factor and O_3 concentration. However, this statistical technique merely reveals the level of correlation between dependent and independent parameters. Furthermore, correlation analysis of only two variables is insufficient to determine the link between more than two variables. However, some factors that cannot account for the third and subsequent factors may exist (Granger, 1969).

Meanwhile, causal relationship analysis is a statistical technique that can help resolve this problem. This method implies a dependency relationship between the cause and effect of each parameter. While the statistical correlation technique reveals only the relationship between parameters, the causal relationship analysis examines the directional cause and effect of each parameter and provides a significant value for the direction. Therefore, the present study used causal relationships to investigate the significant directionality as well as the cause and effect of O_3 concentration on particulate matter pollutants, gaseous pollutants, and meteorological parameters at three different locations in Malaysia, including industrial, urban, and suburban areas. Our findings may provide a useful reference for other researchers and the government to formulate early mitigation measures given the possibility of intensification of anthropogenic activities, which may worsen O_3 pollution.

AIR QUALITY DATA

Secondary air-quality data were obtained from the Malaysian Department of Environment (DoE). These data were continuously recorded and controlled by an automated air quality control remote station while following the established standards required by the Malaysian DoE. From January 2006 to December 2017, the 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, and SO_2) were obtained and converted to monthly average data.

Numerous studies have attempted to explain the fluctuations in O_3 concentrations as a function of gaseous pollutants and volatile organic compound (VOC) precursors; therefore, historical data obtained from the DoE are useful (Ismail et al., 2016; De Souza et al., 2017; Apondo et al., 2018). CO, SO_2, and NO_2 are major gaseous pollutants in the atmosphere that are chemically oxidized to O_3 in the presence of solar light. O_3 and its precursors are transported and accumulated by the wind (Teinilä et al., 2019). High wind speeds may reduce O_3 concentration, allowing pollutants to travel to new locations.

STUDY AREA

Three Malaysian air-monitoring stations were selected for the present study. Peninsular Malaysia comprises various locations and regions. The first air quality monitoring station is located in Nilai (02°15.924′N, E102°10.554′), Negeri Sembilan. The Malaysian DoE classifies this air quality monitoring station as industrial. Nilai air quality monitoring stations are located in rapidly expanding industrial areas with significant air pollution (Ahmat et al., 2015). The second air quality monitoring station classified as the urban type was located in Kota Bharu (06°09.520′N 102°15.059′E), Kelantan. The Kota Bharu air quality monitoring station is located in the northeastern part of Peninsular Malaysia, close to the border with Thailand. Kota Bharu's major activities include trading and tourism (Masseran et al., 2016). The last air quality monitoring station was in Kangar (06°19.545′N 99°51.311′E), the capital of Perlis. It is located in the southern part of Peninsular Malaysia. Suburban areas with extensive human activities are undergoing rapid urban development (Abdullah et al., 2017). The selected air quality monitoring stations in Nilai, Kota Bharu, and Kangar were named S1, S2, and S3, respectively. The geographical map of all three monitoring stations is shown in Figure 1.

Figure 1. Geographical map of the three selected sampling stations

ANALYSIS

The causal relationship statistical technique maximized the cause-and-effect relationship between the dependent and independent parameters considered in the present study. This provided important results regarding the cause and effect of each independent parameter on the dependent parameter, which is reported in terms of significance and direction. The stationarity of each piece of data was confirmed to comply with the causal analysis. Monthly record data for 144 months were utilized. Data on O_3, PM_{10}, gaseous pollutants, and meteorological parameters were subjected to a stationary test. Numerous statistical tests can be used to determine whether the variables in a multivariate time series are stationary.

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

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

where,

α_0: Drift Component

e_t : Independent and homogeneous error terms

To determine the stationarity of the series, Sansudden et al. (2011) have proposed the following hypothesis:

H_0 : The time series data are non-stationary

H_1 : The time series data are stationary

where H_0 is rejected if the significance value (p) is smaller or equal to 0.05.

Thereafter, the causality statistical test was applied using the following equation (Rahmah & Kashem, 2017):

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

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

Subsequently, we tested 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. H_0:b_1 represents the dependent series and H_o:d_1 represents the independent series. Here, a is the coefficient of the series. In each case, rejection of the null hypothesis implies the existence of Granger causality. In other words, Granger causality can be determined using F-statistics and the hypothesis of the Granger causality test is as follows (Jordaan & Eita, 2009):

H_0 : The series is not Granger caused

H_1 : The series is Granger caused

If the significant 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.

Meanwhile, the results of the causal statistical test can be represented graphically. This necessitates a thorough examination of the causes and effects of each parameter. The graphical representation of causal statistics between parameters can be unidirectional or bidirectional. Since a significance test (0.05) was used in the present study, unidirectional indicates that the independent parameter was not a cause and effect of the dependent parameter, or *vice versa*.

Furthermore, if a parameter does not have a Granger cause for O_3 at the significance level of 0.05, it may be significant at a level greater than 0.05. The bidirectionally significant dependent and independent parameters exhibit a relationship of both cause and effect with each other at the significance level of 0.05. Consequently, the direction is significantly affected. Figure 2 shows the details of unidirectional and bidirectional interactions. In the diagram, the dotted lines indicate that parameter B has a cause-and-effect relationship in the direction of the line with parameter A. Simultaneously, the straight line indicates that parameter A does not have a cause-and-effect relationship in the direction of the line with parameter B.

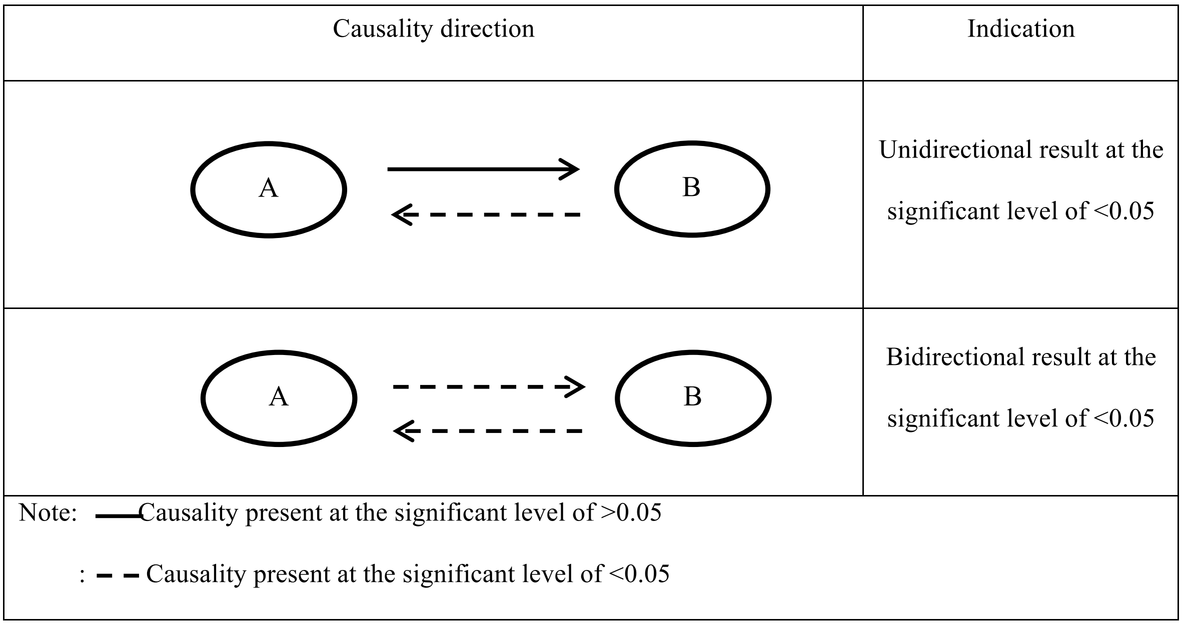

Figure 2. Direction of illustration of causal relationship statistical test

RESULTS AND DISCUSSION

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

The maximum O_3 concentrations recorded at Nilai, Kota Bharu, and Kangar were 0.1140, 0.0830, and 0.0810 ppm, respectively. Thus, O_3 concentration at the industrial sampling station was higher than that that at the urban and suburban sampling stations according to the Malaysian Ambient Air Quality Guideline (MAAQG). The high recorded concentrations of O_3 at the industrial sampling stations were not surprising because of the industrial emissions of NO_X and VOCs as precursors of O_3, which are primarily emitted from industrial processing and heavy transportation activities (Hidy et al., 2015). Furthermore, the disparity in concentrations observed across the three sampling stations may be attributed to differences in local emissions from anthropogenic activities of mobile and stationary sources in terms of atmospheric composition (Banan et al., 2013).

Table 1. Descriptive statistics of O_3 concentrations at the three 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

Hourly averaged data were used to analyze O_3 concentration behavior patterns in depth. The diurnal dispersion of O_3 concentration at the three sampling stations is depicted in Figure 3. At the Kota Bharu and Kangar stations, the O_3 concentration began to rise at 9.00 a.m., peaked at 2.00 p.m., and began to fall at 4.00 p.m. Meanwhile, at the Nilai station, peak concentration was recorded 1 hour later (at 3.00 p.m.) than that at the other two stations, but started to fall at 4.00 pm.

According to the diurnal graph, the industrial area recorded a higher 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 disparities in data. Due to the high intensity of solar radiation (sunlight) in Malaysia, peak O_3 concentrations occur between 1.00 pm and 3.00 pm (Abdullah et al., 2019; Awang et al., 2018). These findings were also confirmed by Geng et al. (2008), who determined that high-intensity solar radiation was the primary contributor to the high recorded O_3 concentrations.

As a result of the different diurnal dispersions of O_3 observed at the three sampling stations, further analysis using the causal relationship statistical technique was performed to determine the parameters that may affect the O_3 concentration. Typically, causal relationship analysis is based on the stationarity of data from records. According to Mills (2015), stationary data are the mean and variance of a dataset that do not change over time.

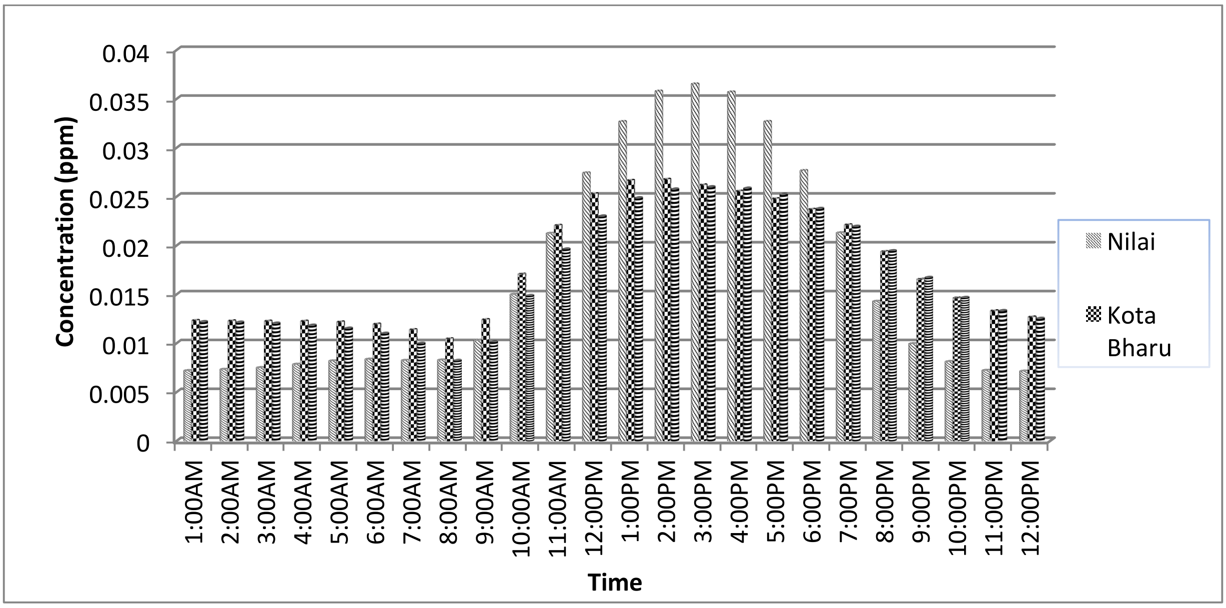

Figure 3. Diurnal dispersion of O_3 concentration at the three monitoring stations

Physical observations using a series plot graph may not always provide clear information regarding the stationarity of each set of recorded data. Therefore, the ADF test was run on all datasets for the three monitoring stations to examine stationery. The ADF test was suitable for use to check the stationarity of our data, and the null hypothesis was rejected at the given level of confidence (Omar et al., 2013).

Table 2 summarizes the t-statistics and p-values of the ADF test on nonstationary results for the recorded air pollutant and meteorological data. According to the ADF test, all parameters at the Nilai station had non-stationary data records. Furthermore, the sampling stations in Kota Bharu and Kangar had six and five stationary parameters, respectively. Temperature and relative humidity were nonstationary parameters in Kota Bharu. At Kangar, ground-level ozone, nitrogen dioxide, and relative humidity were recorded as nonstationary data. Given the non-stationarity of several parameters, differencing was required to convert the data to a stationary dataset to comply with the rules of causality methodologies.

Table 3 presents the results of causality relationships. Three parameters, namely SO_2, NO_2, and WS, showed a causal relationship with O_3 concentration in Nilai at the significance level of <0.05, with p-values of 0.0132, 0.0086, and 0.0475, respectively. Meanwhile, SO_2 and CO in Kota Bharu and NO_2 and CO in Kangar showed a causal relationship with O_3 concentration at the significance level of 0.05.

Table 2. ADF statistics and p-values for non-stationary dataset of the three monitoring stations

Station	Parameter	t-statistics	p-value
Nilai	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	T	-0.4819	0.6795
	RH	-0.5151	0.1079
Kangar	O_3	-1.4860	0.1280
	NO_2	-1.2538	0.2068
	RH	-0.3135	0.6065

Moreover, detailed cause and effect relationships between parameters in the directional causality were obtained. The results of directional causality test are the major findings that distinguish the present study from the previous correlation studies on air quality. Figure 4 depicts the outcomes of the directional parameters. The causality test was found to be significant at the 0.05 level, because no bidirectional events occurred. At the significance level of <0.05, one-directional causality was noted between O_3 and NO_2, SO_2, and WS at the Nilai sampling station. Furthermore, SO_2 and CO were found to be causal parameters for O_3 in Kota Bharu, whereas NO_2 and CO were found to be the causal parameters at Kangar.

As shown in Figure 4, three parameters showed a cause and effect at the significance level of 0.5 at Nilai and Kota Bharu. Furthermore, the cause-and-effect relationship between SO_2 and NO_2 at Nilai was bidirectional. At the significance level of 0.05, the causal relationship analysis revealed that air pollutant concentrations and meteorological parameters showed varied causation relationships at different monitoring stations. However, as the precursors of O_3 pollutants, such gaseous pollutants as SO_2, NO_2, and CO may have a causal relationship with O_3.

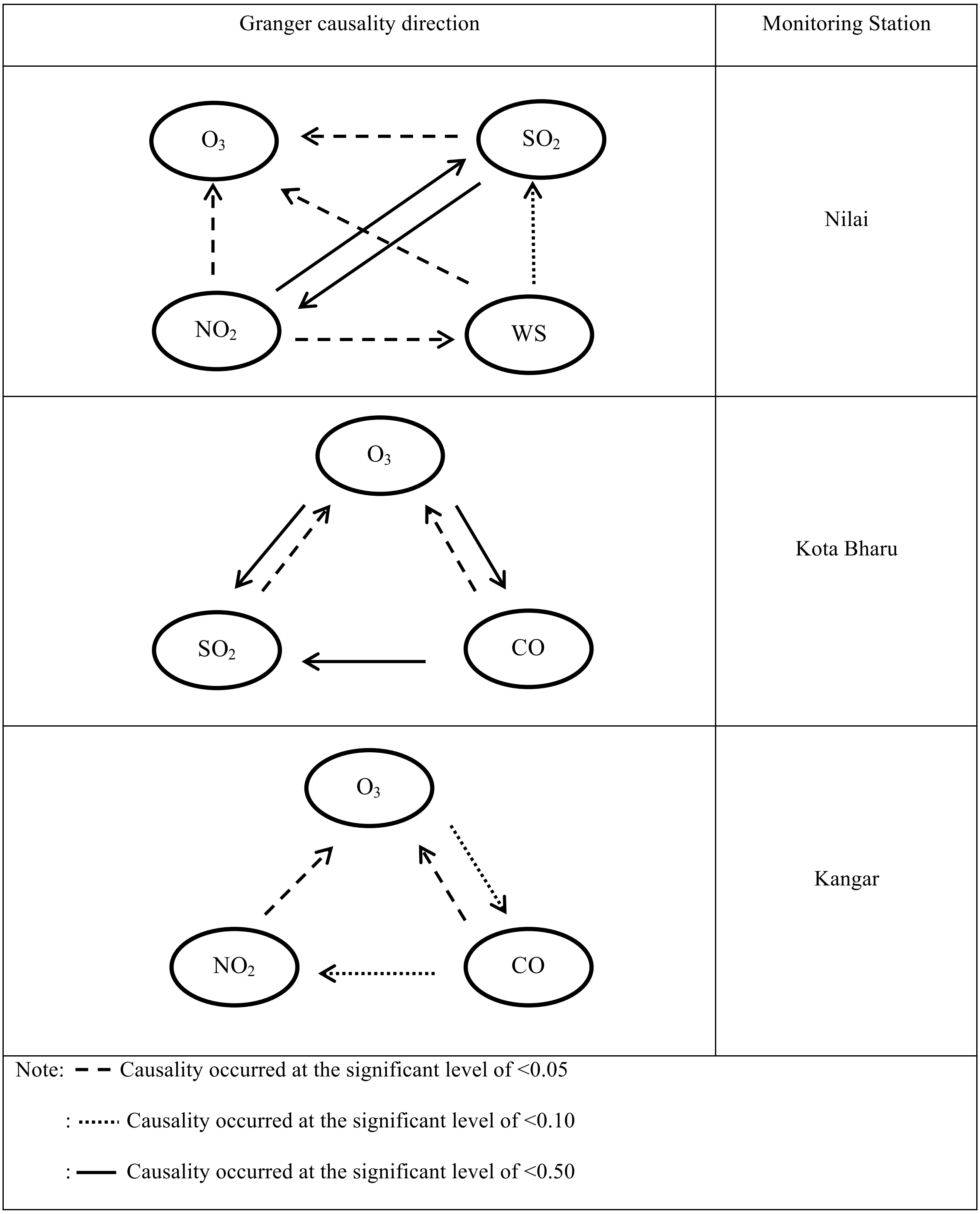

Figure 4. Direction of causality at all monitoring stations

The results of causality relationship in the industrial area (Nilai) were expected due to regional and local emissions, which refer to transport and motor vehicle emissions, respectively. Meanwhile, in the urban (Kota Bharu) and suburban (Kangar) areas, the results were expected due to regional and local emissions, which refer to transport and motor vehicle emissions, respectively. Simultaneously, the transfer of wind emissions from nearby places, such as industrial areas, to urban or suburban areas, causes significant O_3 pollution.

Table 3. Causality relationship t-statistic and *p*-values for all sampling stations

Nilai							
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							
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							
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
Parameters affecting O_3 concentration at the significant level of <0.05 are indicated in bold							

CONCLUSION

Our findings indicate that industrial (Nilai) monitoring stations recorded higher O_3 concentrations than urban (Kota Bharu) and suburban (Kangar) monitoring stations. Moreover, the maximum O_3 concentrations that exceeded the acceptable limit of the MAAQG were only found in Nilai. Meanwhile, in Nilai, Kota Bharu, and Kangar, the diurnal dispersion of O_3 concentration followed a similar trend. However, O_3 concentration peaked at midday, following a rapid rise in the morning, and then gradually declined to a low level in the evening. The results of the causal association between O_3 pollutants and other parameters (e.g., PM_{10}, SO_2, NO_2, CO, T, WS, and RH) indicated that gaseous pollutants, such as SO_2, NO_2, and CO, showed a causal relationship with O_3 concentration at the significance level of <0.05. In Nilai, wind speed was the sole meteorological parameter showing a causal relationship with O_3 at the significance level of 0.05. However, all causal relationships at the three selected monitoring stations examined were unidirectional, the bidirectional causal relationship of O_3 with SO_2 and NO_2 at the significance level of not more than 0.5 at Nilai. This finding was expected owing to the mechanism of O_3 pollutant, which requires these parameters to build up in the ambient air. Overall, the parameters affecting O_3 concentration clarified using the causal relationship analysis in the present study can offer an important reference for other researchers to improve air quality studies, particularly for prediction purposes.

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