



The Influence of Spatial Variability of Critical Conversion Point (CCP) in Production of Ground Level Ozone in the Context of Tropical Climate

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ABSTRACT

Critical conversion point (CCP) is a very crucial step in production of the ground level O₃ chemistry. Thus, a multivariate analysis was applied on the dataset of nine selected locations in Malaysia from 1999 to 2010. It incorporated hierarchical agglomerative cluster analysis (HACA) to explore the spatial variability of CCP and principal component analysis (PCA) to determine the major sources of the air pollutants that influence ozone CCP. High variability in CCP was observed between the monitoring stations that occurred during critical conversion time (CCT) from 8:00 a.m. to 11:00 a.m. The HACA results grouped the nine monitoring stations into three different clusters, based on the characteristics of ozone concentrations during CCT period. Results of PCA for the three clusters showed that the contributions to O₃ level variation during CCT by meteorological variables (UVB, temperature, relative humidity, and wind speed) are higher at 51.6%, 48.5%, and 33.3% than that of primary air pollutants (NO₂, SO₂, PM₁₀) at 19.2%, 21.4%, and 15.2% for cluster 1, cluster 2, and cluster 3, respectively. Therefore, applying a targeted spatial control strategy for ground level O₃ precursors during the CCT period is a crucial step.

Keywords: NO₂ photolysis; NO titration; Critical conversion point; Multivariate analysis.

INTRODUCTION

Ground-level ozone (O₃) is one of the criteria air pollutants that is always associated with degradation of air quality worldwide. It induces harmful effects on human health, crop production, material quality, and the ecosystem. As a secondary air pollutant that is produced from anthropogenic activities, the formation and accumulation of O₃ are induced by the emissions of nitrogen oxide (NO_x) and volatile organic compounds (VOCs) (Seinfeld and Pandis, 2006). O₃ formation is very responsive to changes in meteorological parameters. Thus, elevated O₃ levels are often associated with intensive solar radiation, high temperature, minimal rainfall, low wind speed, and low relative humidity (Toh *et al.*, 2013).

The dependency of O₃ formation toward UV light causes its clear daily variations. In the presence of sunlight, nitrogen dioxide (NO₂) undergoes photochemical reactions to produce free oxygen atom (O), which later reacts with oxygen molecules (O₂) to form O₃ (Duenas *et al.*, 2004; Azmi *et al.*, 2010). Once O₃ is created, it is destroyed through

several pathways, such as nitric titration and surface deposition (Abdul-Wahab *et al.*, 2005). O₃ concentration variations show an interesting patterns in the morning where O₃ level reaches the lowest concentration because of the higher rate of NO titration (Jiménez-Hornero *et al.*, 2010). Once the minimal point is reached, O₃ starts to increase with rising NO₂ concentration, thereby promoting NO₂ photolysis. When the NO₂ photolysis rate is higher than the NO titration rate, critical conversion point (CCP) occurs. Therefore, CCP is very crucial step in ground-level O₃ chemistry because the different in the chemical reaction's rate is expected to result in O₃ accumulation.

The background O₃ has increased over the last decade and is expected to continuously increase in the subsequent years (Ghosh *et al.*, 2013). Thus, many countries, including Malaysia, monitor the current O₃ condition and have set guidelines against this air pollutant. In 2010, the Recommended Malaysian Air Quality Guideline (RMAQG) of 100 ppbv for the hourly O₃ is often exceeded in several places such as at monitoring stations in Klang Valley (Latif *et al.*, 2012) which imperil health problems and ecological impact for millions of people lived in this region. The study of ozone variations is complex because of various possible precursors, photochemical processes, and meteorological factors (Chattopadhyay and Chattopadhyay, 2011; Toh *et al.*, 2013). In addition, the interactions among O₃, its precursors, and meteorological parameters occur within a wide range

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of temporal and spatial scales (Abdul-Wahab *et al.*, 2005). Thus, implementing a targeted control strategy by location, such as a particular place, and by time, such as the morning rush-hour time period is crucial step to minimize O₃ precursors' emission reductions. Therefore, this study is attempted to introduce the possibilities to use CCP in explaining the production of the ground level O₃ and to explore the spatial variability of CCP in the context of tropical climate.

METHODS

Study Areas Description

The dispersion and dilution of air pollution are directly influenced by local attribution such as meteorological condition as well as the location of the monitoring stations

(Hosseiniabalam *et al.*, 2010). Thus, nine continuous air quality monitoring stations operated by Alam Sekitar Malaysia Sdn Bhd (ASMA) was selected in this study and covered three different types of land use: industrial (Pasir Gudang, Perai, Kemaman and Kuching), urban-residential (Kota Bharu, Kota Kinabalu, Gombak and Klang), and one reference site (Jerantut). All the stations except Jerantut are highly polluted due to industrial activity, traffic emissions and rapid development by growing populations in these areas. Fig. 1 shows the map of the study area and Table 1 shows the description of the selected monitoring stations.

Perai monitoring station (PR) is located in Seberang Perai Penang, one of the most heavily populated states in Peninsular Malaysia. The main air pollution emission contribution comes from industrial and traffic emission. In addition, power plants can also be considered as possible



Fig. 1. Specific location of monitoring stations across Malaysia (map is not up to scale).

Table 1. Description of monitoring selected monitoring stations.

Group	Stations	Station ID	Locations	Coordinate	Area (km ²)	*Population
Industrial	Pasir Gudang	PG	South	N01°28.225 E103°53.637	311.3	46,571
	Kemaman	KE	Middle	N04°16.260 E103°25.826	2,581	173,000
	Perai	PR	North	N05°22.265 E100°23.344	738	362,820
	Kuching	KC	South	N01°33.734 E110°23.329	1863	600,200
Urban	Kota Bharu	KB	North	N06°09.520 E102°15.059	403	509,400
	Kota Kinabalu	KK	North	N05°53.623 E116°2.596	351	423,300
	Gombak	GB	Middle	N03°15.924 E101°39.103	628	681,300
	Klang	KL	Middle	N03°00.620 E101°24.484	636	832,600
BG	Jerantut	JT	Middle	N03°58.238 E102°20.863	7,241	98,300

Note: *2010; BG is background.

sources of ozone precursors' since there are three power plants are stated within 25 km in the study area. Gombak (GB) and Klang (KL) monitoring stations are located in Selangor, the most developed and heavily populated state in Malaysia. Klang is one of the districts in Selangor that was known as a port city. Meanwhile, Gombak monitoring station is surrounded by high-populated residential areas and major roads which experiences heavy traffic during morning and evening rush hour (Azmi *et al.*, 2010). Pasir Gudang monitoring station (PG) is situated in Johor state (Southern State of Peninsular Malaysia) where Pasir Gudang Port is the fourth busiest port in Malaysia (DoSM, 2012). In 2010, Port of Pasir Gudang handled about 28 million tons of cargo and received nearly 5000 international and local vessels (MoT, 2012). The major industries that drive the economy of Pasir Gudang are transportation and logistics, shipyard industries, petrochemical industries, as well as oil palm storage and distribution. Kemaman is situated in Terengganu (East Coast of Peninsular Malaysia), the city is relatively less developed, except a few places along the coastline where steel and petroleum plants are located (Sulong *et al.*, 2002).

Kota Bharu monitoring station was located in Kelantan. According to Shaari *et al.* (2012), the major land use in Kota Bharu is for agriculture, with one industrial park located at Pengkalan Chepa (Azlan *et al.*, 2011). Kuching monitoring station is located at northeast Borneo in Sarawak state which is surrounded by industrial activities. Besides that, this station is also affected by power plants such as PPLS Power Generation Plant (coal-fired) and Sejingkat power Corporations Plant (coal-fired) (Chung *et al.*, 2012; Dominick *et al.*, 2012). Kota Kinabalu monitoring station is in Kota Kinabalu, the capital city of Sabah state (North Borneo), the station is surrounded by high-populated residential areas and major roads. Jerantut as a background station is located at MMS (Meteorology Monitoring Station), Batu Embun. The station is surrounded by the agricultural area and traditional Malaysian villages (Banan *et al.*, 2013). According to Azmi *et al.* (2010), the source of the air pollution in Jerantut is expected to be natural forest fires, open burning, soil dust, and motor vehicles.

Weather Condition of Study Area

Climatically, Malaysia experience tropical rainforest climate distinguished by high temperature ranging from 22 to 24°C during night time and from 27 to 30°C during daytime. Seasonal variations in Malaysia are distinguished by changes in wind flow patterns and rainfall intensity (Md Yusof *et al.*, 2010). Uniform periodic changes in wind flow patterns and rainfall intensity are described as monsoonal changes. Peninsular Malaysia has two monsoonal seasons per year, which are the northeast monsoon (NEM) (November–March) and the southwest monsoon (SWM) (June–September), and two intermonsoon period occurred during April to May and October to November. The mean annual rainfall in these locations is approximately 2670 mm (Ghazali *et al.*, 2010; Md Yusof *et al.*, 2010), and the relative humidity ranges from 70% to 90%. Heavy seasonal rains observed during northeast monsoon (November to

January) (Sulong *et al.*, 2002), while the driest months are June and July.

Monitoring Records

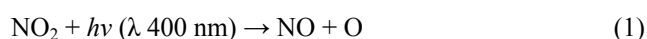
Continuous hourly ground level O₃ concentrations and other air pollution levels were established across Malaysia by Department of Environment for measuring and detecting any significant changes in air quality from 1999 to 2010. Hourly O₃ concentrations are measured using the UV absorption O₃ Analyzer Model 400A, which is a microprocessor controlled device (Mohammed *et al.*, 2013). The O₃ analyzer applies a system based on the Beer-Lambert law to measure low ranges of O₃ concentration in ambient air and gaseous media (Ghazali *et al.*, 2010). Ambient O₃ concentration is detected from internal electronic resonance of O₃ molecules using absorption of 254 nm UV light emitted from an internal mercury lamp (Teledyne, 2011). Meanwhile, hourly NO₂ and NO concentrations were collected using the NO/NO₂/NO_x analyzer model 200A (Ghazali *et al.*, 2010). The analyzer applies the chemiluminescent detection principle to detect NO₂ concentration in ambient air. The procedures employed were adopted from the standards outlined by internationally recognized environmental organizations such as the United State Environmental Protection Agency (USEPA) (Latif *et al.*, 2014). Meanwhile, incoming solar radiation was measured based on ultraviolet beta (UVB) rays with wavelength ranging from 280 nm to 315 nm. Hourly average temperature, relative humidity and UVB vibrations were measured with Met One 062 sensor, Met One 083D sensor, and Scintec Model UV-S-290-T, respectively. The number of each air pollutants and meteorological parameters observations in this study totals 157,680 (1461 observation per parameter × 9 stations × 12 years). The descriptive statistics of the measured 12 year data set are summarized in Table 2.

Determination of CCP Using Composite Diurnal Plot

Graphically, CCP were determined based on composite diurnal plots of O₃, NO₂, NO, temperature and UVB radiation. CCP was assuming to occur at point of intersection between O₃, NO₂, and NO diurnal plots line. If the exact intersection point cannot be obtained from the plots, estimation of the interception point was point out as the CCP.

Determination of Critical Conversion Time (CCT) Based on Ozone Production Rate

At the ground level, it is established that there is inter-conversion between O₃, NO₂, and NO concentration that is dominated by reactions as follows (Ghazali *et al.*, 2010):



With the availability of photons with wavelength shorter than 424 nm, the photo stationary state in which the concentration of NO₂ and NO were related to the O₃

Table 2. Descriptive statistics of daily average of air pollutants and meteorological factors from 1999–2010.

Stations	O ₃ (ppb)			NO ₂ (ppb)			NO (ppb)			T (°C)			UVB (J m ⁻² hr ⁻¹)		
	Mean	SD	Max	Mean	SD	Max	Mean	SD	Max	Mean	SD	Max	Mean	SD	Max
PG	10.9	8.86	79.0	13.1	7.2	57.0	23.3	28.2	266.0	26.8	2.5	38.9	175.7	171.6	903.0
PR	10.6	9.27	76.0	14.3	6.4	59.0	23.0	19.8	165.0	27.0	2.8	37.2	175.1	175.7	855.0
KC	8.7	6.66	46.0	7.4	5.1	56.0	11.4	8.5	165.0	27.0	3.3	36.9	249.2	222.7	1035.0
KE	15.6	10.5	63.0	4.9	7.6	72.0	2.7	5.2	212.0	26.3	3.2	38.6	164.3	161.2	703.0
KB	10.9	8.2	63.0	6.5	4.0	106.0	9.1	9.7	106.0	26.4	2.6	35.7	211.8	184.8	843.0
KK	13.9	8.7	55.0	3.7	2.5	22.0	5.5	5.8	72.0	28.8	2.7	37.4	334.9	247.2	1159.0
GB	9.5	10.4	75.0	17.1	10.4	197.0	48.6	43.2	316.0	26.6	3.3	38.7	157.7	160.2	747.0
KL	12.5	10.7	72.0	23.2	11.0	99.0	33.5	33.9	329.0	27.9	3.0	37.8	166.7	170.0	886.0
JT	8.7	6.8	50.0	2.1	1.3	18.0	4.7	25.6	550	25.1	2.5	35.6	166.2	164.8	747.0

concentration is given by the follows Eq. (4) (Seinfeld and Pandis, 2006):

$$\frac{d[\text{NO}_2]}{dt} = -j_{\text{NO}_2} [\text{NO}_2] + k_3 [\text{O}_3] [\text{NO}] \quad (4)$$

Assuming that [O] is not constant and rather varies with [NO₂] and consequently achieved instant balance between its rate of production and loss. According to the reactions before, there is a point where NO₂ is destroyed and reproduced at a very fast rate that will induced a steady-state cycle is maintained (Seinfeld and Pandis, 2006). The steady-state ozone concentration is given by the follows Eq. (5) (Notario *et al.*, 2013):

$$[\text{O}_3] = \frac{j_{\text{NO}_2} [\text{NO}_2]}{k_3 [\text{NO}]} \quad (5)$$

where j_{NO_2} is the rate of NO₂ photolysis; k_3 is the rate of NO titration. j_{NO_2}/k_3 was used to indicate the variations the rates of NO₂ photolysis and NO titration. The positive differences of j_{NO_2}/k_3 rates with the previous hour indicating that NO₂ photolysis rates higher than NO titration rates, while negative indicate that NO₂ photolysis were lower than NO titration. The biggest positive differences were used to point out the time for CCP.

Statistical Analysis

In this study, continuous data of selected variables are analyses according to spatial location. The mean, median, minimum, maximum and standard deviations of each variable are calculated to overview the distribution of data. For this study, no imputations methods were applied and any missing values occurred during data acquisition were omitted from analysis.

Hierarchical Agglomerative Cluster Analysis

Hierarchical agglomerative cluster analysis (HACA) is a set of multivariate techniques commonly used to group the objects into clusters so that the objects (monitoring stations) within a cluster are similar to each other while objects located in other clusters are different from each other. HACA maximizes the similarity of cases within each cluster while

minimizing the dissimilarity between groups that are initially unknown (Lu *et al.*, 2011). In this analysis, each object is considered as a separate cluster before it is connected by Ward method agglomerate techniques and squared Euclidean distance to measure the similarity between hourly ozone concentrations using Eq. (6). Wards method is chosen as the linkage in this study because this method used an analysis of variance approach in evaluate the distance between clusters in attempt to minimize the sum of square (SS) of any two cluster (Shrestha and Kazama, 2007). The classification of the objects can be illustrated in a dendrogram (tree diagram), which shows the measured similarity or distance between any two variables.

$$d_{ij}^2 = \left[\sum_{k=1}^p (X_{ik} - X_{jk})^2 \right] \quad (6)$$

Hierarchical cluster analysis can be cut at any level. The optimum number of clusters is usually determined using the difference in distance values as the optimum point is where clear declamation between differences in distance is recorded. Then, CA is repeated using the selected number of cluster to evaluate the selected number of optimum cluster. Further, to accommodate practicality of the results as there is ample information $D_{\text{max}}/D_{\text{link}} \times 100 < 15$ step is used (Shrestha and Kazama, 2007).

Principal Component Analysis (PCA)

PCA is a multivariate technique that is widely used to deal with voluminous data in monitoring studies, such as air pollution research. In the present study, this technique was applied to reduce variables and to identify the most relevant variables in O₃ variations (Dominick *et al.*, 2012). PCA is widely known due to its capability to detect the most significant variables in dataset with minimum loss of the original information (Dominick *et al.*, 2012; Elbayoumi *et al.*, 2014). Principal components (PCs) were extracted, such that the first PC (PC1) accounted for the largest amount of total variation in the data set, whereas the following components accounted for the remaining variations that were not considered in PC1 (Kovac-Andric *et al.*, 2009). PCs are generally expressed as follows:

$$PC_i = l_{1i}X_1 + l_{2i}X_2 + \dots + l_{mi}X_m \quad (7)$$

where PC_i is the i^{th} principal component, and l_{mi} is the loading of the observed variable X_m

The significant variables for each component are determined based on the loading. In this study, only a factor loading that is greater than 0.4 is considered significant (Ul-Saufie et al., 2013). The sufficiency of the monitoring data for PCA was assessed using Kaiser-Meyer-Olkin (KMO) and Bartlett's tests. According to Özbay et al. (2011), these tests are applied to examine the hypothesis that the variables are uncorrelated in the population. The KMO result (0.735) showed that the value was greater than 0.5, which indicated that the data were sufficient for PCA. Meanwhile, for

Bartlett's Test of sphericity (35524.2) showed that the selected variables were significantly ($p > 0.001$) related to one another and suitable for factor analysis.

RESULT AND DISCUSSION

Critical Conversion Point Based on Composite Diurnal Plots

The annual CCP at a locations were determined based on the composite diurnal plot of O_3 , NO , NO_2 concentrations, temperature and UVB in depicted in Figs. 2, 3 and 4. It is observed that, CCP at the studied locations ranged from 8

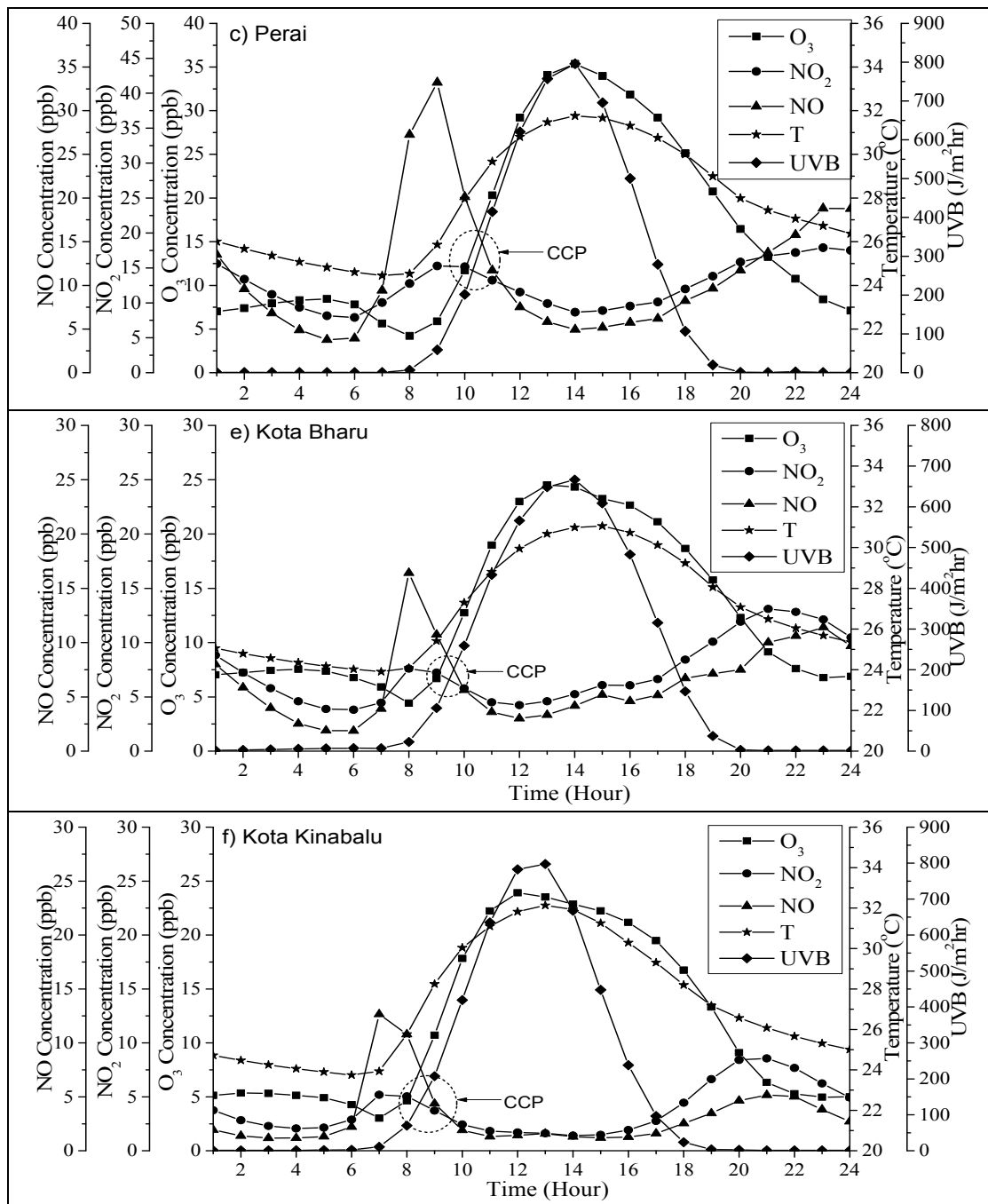


Fig. 2. CCP based on composite diurnal plots for monitoring station located in north Malaysia.

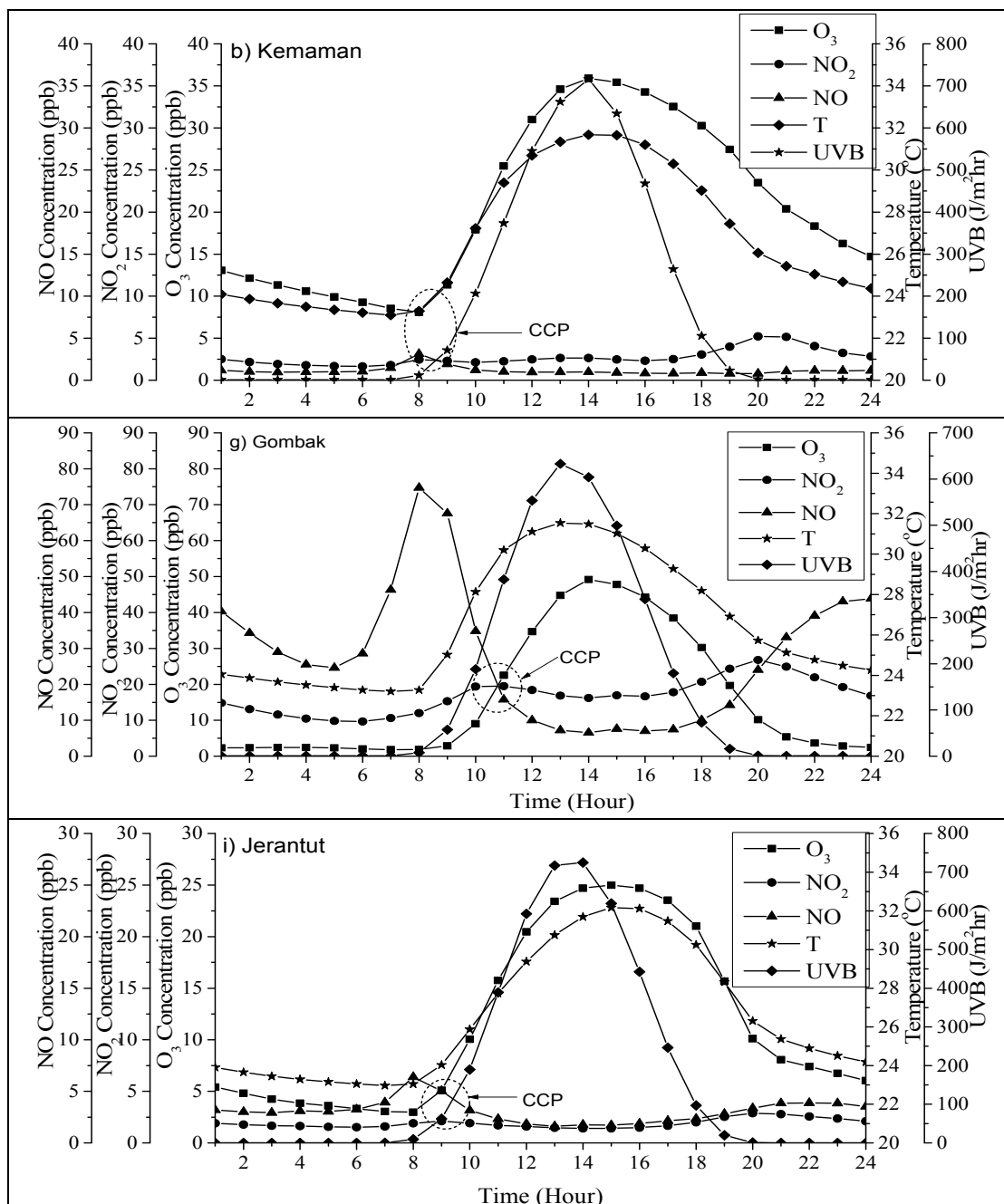


Fig. 3. CCP based on composite diurnal plots for monitoring station located in middle Malaysia.

a.m.–11 a.m. The earliest time was measured at Kota Kinabalu and followed by Kemaman with all recorded CCP were around 9 a.m. At most of the industrial stations, the CCP were constantly measured around 9 a.m.–10 a.m. Furthermore, the result suggested that, at most of the urban station, CCP were around 10 a.m.–11 a.m. except for Kota Kinabalu. Earlier time for CCP Kota Kinabalu, Kuching and Kemaman were expected due to the earlier sunrise at these locations. At Jerantut, CCP is approximately determined at \pm 9 a.m., since the exact intersection between O_3 , NO_2 and NO concentration is could not be obtained. Since the CCP is determined based on the composite diurnal plot of O_3 , NO_2 , NO , T and UVB , these variables are the factors

that controlled time of CCP occurrences. In the presence of sunlight, the NO_2 photolysis was able to complete and accumulated as the level of precursors increasing. However, at urban stations such as Gombak and Klang the CCP were later than at other places contributed to higher NO concentration during early morning (6 a.m.–8 a.m.).

CCP is assumed as the interception point between O_3 , NO_2 , and NO line in the composite diurnal plots. The plots illustrated that after reached the CCP, O_3 will showed increment trends, while NO_2 and NO will showed decrement trends. It is observed that, O_3 concentrations in stations that their CCP occur around 10 a.m. and later are relatively higher than in station that have their CCP earlier. The mean

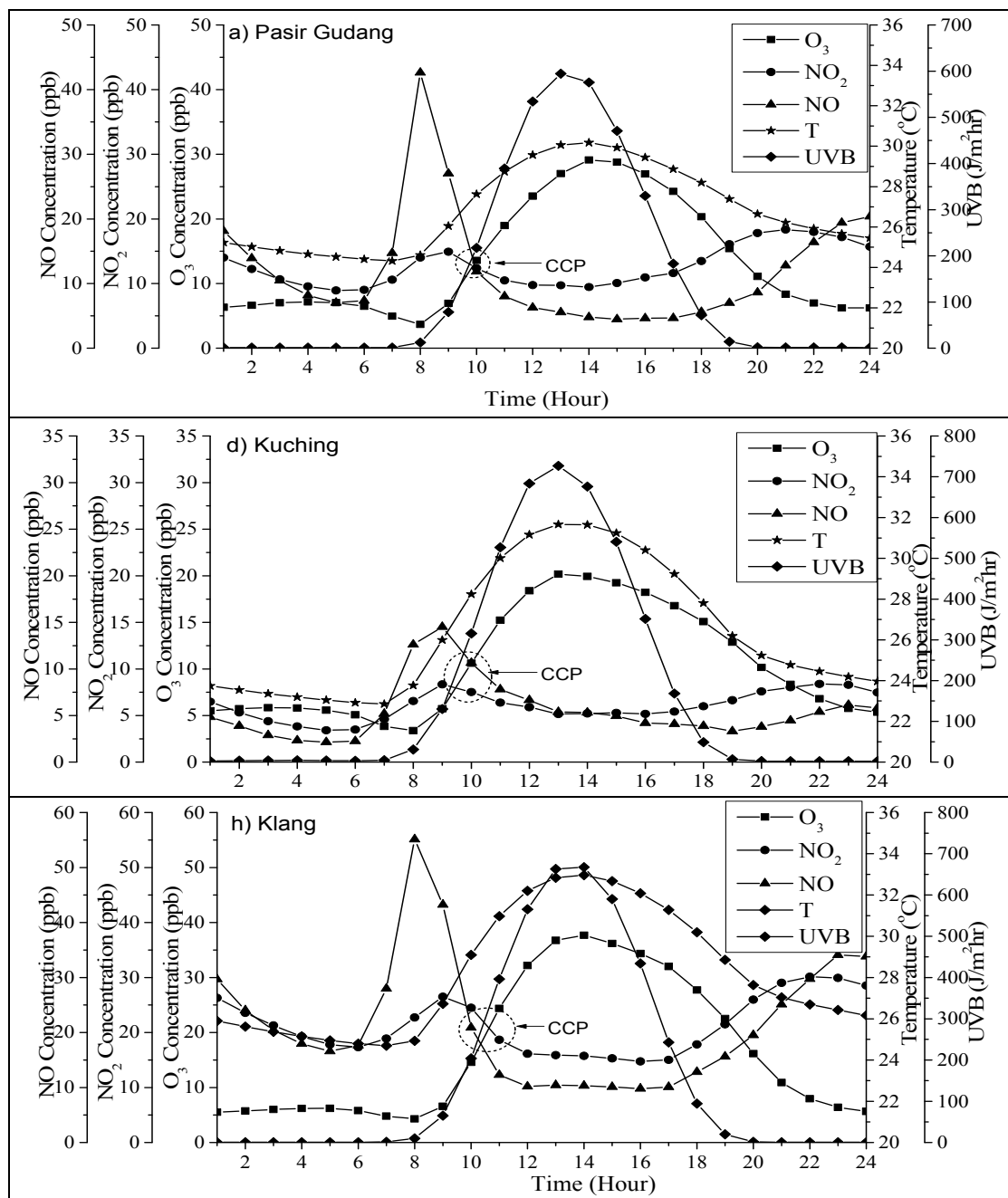


Fig. 4. CCP based on composite diurnal plots for monitoring station located in south Malaysia.

O₃ concentration for Klang and Gombak is around 20 ppb, while for Kota Kinabalu, Kota Bharu and Jerantut is around 10 ppb. This finding opens the possibilities to use time for CCP to give some indication regarding O₃ concentrations level at one particular location. According to Reddy *et al.* (2011) and Jiménez-Hornero *et al.* (2010), the NO concentration plays an important role in the reduction of ozone concentration in the morning. From 6 a.m. to 8 a.m., high concentrations of NO are released by anthropogenic sources such as motor vehicles and industrial establishments. Due to high concentration of NO, the NO titration rate will also increase, hence increase O₃ scavenge which will profoundly reduce O₃ concentrations. The minimum O₃

concentrations in Klang, Gombak are 4.31 ppb and 1.82 ppb, respectively. However, high NO titration will ultimately produce high concentration of NO₂, which is later converted into O₃. In these stations, the time for CCP will be later because NO₂ photolysis rate will take longer to surpass NO titration. Alternatively, the time for CCP will be faster when NO titration rate is lower.

The annual variations of CCP time were expected to occur due to the variation in the intensity of precursor's concentration. Fig. 5 illustrates the CCP during 1999–2010 based the annual average composite diurnal plots based on the differences in stations type. High variability in CCP was observed at Kemaman with the magnitudes of differences

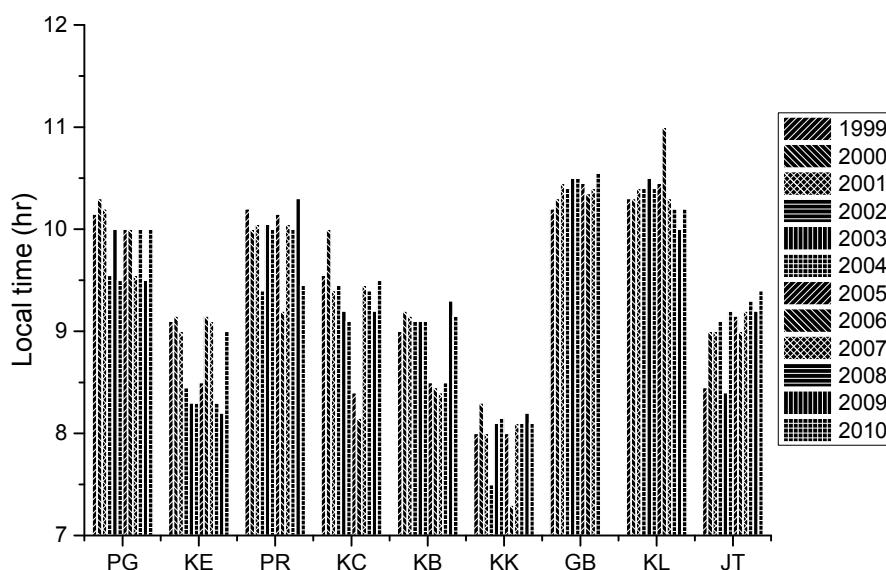


Fig. 5. The annual critical conversion time of ozone formation at all station based on composite.

between the earliest and latest time is around 40 minutes. Result suggested that, latest time for CCP were observed at Klang during 2006. In general, at most of the studied monitoring stations, CCP was occurred at approximately between 9 a.m. and 10 a.m. Further, the majority of stations that were CCP measured at ± 9 a.m locate at east coast of Malaysia. Kemaman showed characteristically difference in CCP from other locations since at the station the concentration level of NO_2 and NO is at very low. Further, only small variation in time for CCP as constant time is observed for each year in all stations. Due to this, the effect of weather conditions is considered very minimal and hardly effected the CCP variations. In addition, the usage long period of data (12 years) influence the effect of weather condition.

Critical Conversion Point Based on Photochemical Reactions

Table 3 shows the calculated value of j_{NO_2}/k_3 at industrials, urban and background stations, respectively. Jenkin and Clemitshaw (2000) reported that as a result of rapid inter-conversion, the behaviour of NO and NO_2 is highly coupled. Thus, in the absence of any competing inter-conversion reactions at the ground level, a photostationary state of the relations between O_3 , NO and NO_2 is obtained through Eqs. (1) to (5). The j_{NO_2}/k_3 value fluctuates throughout the day as the concentration of O_3 , NO_2 and NO also varies daily. Result in the table suggested that, the daily variations of j_{NO_2}/k_3 value are similar to O_3 daily variations. Han *et al.* (2011) also reported similar findings when calculating the j_{NO_2}/k_3 at Tianjian, China. Theoretically, the j_{NO_2}/k_3 value is supposed to be zero during the night time in absence of the photochemical reactions, however the background O_3 concentration in the atmosphere cause the j_{NO_2}/k_3 is at minimal around 5–7 ppb. The ratio value starting to increase during daytime and reaching maximum value at 2 PM around 20 ppb, coincide with the maximum daily O_3 concentration that were observed at most of the studied monitoring

stations. After reaching the maximum point, the j_{NO_2}/k_3 value is decreasing as the NO_2 concentration were gradually increased reaching its evening peaks that often measured at 9 p.m.–10 p.m.

The differences in j_{NO_2}/k_3 value at current hour (h_i) to the previous hour (h_{i-1}) denoted as $\Delta j_{\text{NO}_2}/k_3$ used as indicators of the differences of the rates of NO_2 photolysis and NO titrations (Clapp and Jenkin, 2001). This value was used as indicators to different in rate of NO_2 photolysis and NO titrations and determination of CCP when the biggest positive $\Delta j_{\text{NO}_2}/k_3$ value occurred. High positive $\Delta j_{\text{NO}_2}/k_3$ value will lead to O_3 accumulation, while high negative $\Delta j_{\text{NO}_2}/k_3$ value is expected to contribute to O_3 destruction. The result in Table 3 exhibited that, the largest $\Delta j_{\text{NO}_2}/k_3$ is measured as early as 7 a.m. at Kota Kinabalu and as latest as 12 pm measured at Klang. Similar to CCP, the highest $\Delta j_{\text{NO}_2}/k_3$ mostly occurred at 10 a.m. At several locations such as Kajang and Gombak the time for CCP is coincided with $\Delta j_{\text{NO}_2}/k_3$ time. Meanwhile, at other locations, the CCP and $\Delta j_{\text{NO}_2}/k_3$ is apart by an hour differences, which is $\Delta j_{\text{NO}_2}/k_3$ were measured at an hour earlier and an hour later than the CCP time. However, at Kota Kinabalu, Jerantut, Pasir Gudang and Johor Bahru, the time for CCP and $\Delta j_{\text{NO}_2}/k_3$ is minimally 2 hour apart. Technically, the calculations of $\Delta j_{\text{NO}_2}/k_3$ can be used as the aided techniques in finding the CCP of O_3 formations at a certain locations.

HACA

HACA was performed on O_3 levels during the CCT period which was from 9 a.m. to 11 a.m. to study the spatial variations of the air monitoring stations based on their similarity levels over the period of 1999 to 2010. Nine monitoring stations (i.e., Pasir Gudang, Perai, Kemaman, Kuching, Kota Bharu, Kota Kinabalu, Gombak, Klang and Jerantut) were applied to Hierarchical Agglomerative Cluster Analysis (HACA). The nine stations were classified into three clusters (Fig. 6). Cluster 1 was formed with KC, JT, PG, KB, KK and PR. Cluster 2 took GB and KL, while

Table 3. The calculated value of j_{NO_2}/k_3 for monitoring stations.

Time	KL		JB		PG		KB		KK		JT		KE		KC		GB	
	a^*	b^*	a^*	b^*	a^*	b^*	a^*	b^*	a^*	b^*	a^*	b^*	a^*	b^*	a^*	b^*	a^*	b^*
1	6.2	-0.5	9.1	-1.1	8.2	0.1	6.4	0	2.6	-0.2	9.1	-1.1	6.14	0.11	4.1	0	6.3	0
2	5.9	-0.3	8.2	-0.9	7.5	-0.7	5.9	-0.5	2.6	0	8.2	-0.9	5.7	-0.44	4.2	0.1	6.1	-0.2
3	5.8	-0.1	7.5	-0.7	6.9	-0.6	5.1	-0.8	2.7	0.1	7.5	-0.7	5.66	-0.04	3.8	-0.4	6.1	0
4	5.8	0	7.3	-0.2	6.1	-0.8	4.2	-0.9	2.9	0.2	7.3	-0.2	5.89	0.23	3.5	-0.3	5.9	-0.2
5	5.8	0	7	-0.3	5.6	-0.5	3.6	-0.6	3	0.1	7	-0.3	5.88	0	3.5	0	5.8	-0.1
6	5.9	0.1	7.1	0.1	5.3	-0.3	3.3	-0.3	3.3	0.3	7.1	0.1	5.89	0.01	3.3	-0.2	6	0.2
7	7.1	1.2	7.5	0.4	6.9	1.6	5.2	1.9	7.4	4.1	7.5	0.4	6.91	1.02	4.3	1	7.9	1.9
8	10.5	3.4	10	2.5	11.2	4.3	9.6	4.4	9.9	2.5	10	2.5	10.31	3.4	6	1.7	11.6	3.4
9	10.8	0.3	12.1	2.1	12.6	1.4	9.9	0.3	12.6	2.7	12.1	2.1	9.5	-0.81	9.9	3.9	12.7	1.1
10	12.4	1.6	16.5	4.4	13	0.4	12.4	2.5	14	1.4	16.5	4.4	10.22	0.72	15	5.1	16.2	3.5
11	16.1	3.7	21.5	5	14.5	1.5	15.2	2.8	16	2	21.5	5	11.58	1.35	18.6	3.6	18.2	2
12	20.3	4.2	23.7	2.2	15.1	0.6	16.4	1.2	20	4	23.7	2.2	12.11	0.53	20.7	2.1	18.8	0.6
13	24.2	3.9	25.6	3.2	15.4	0.3	17.9	1.5	23.1	3.1	25.6	1.9	12.72	0.61	21.1	0.4	19.1	0.3
14	24.7	0.5	31	4.1	14.8	-0.6	19.3	1.4	21.2	-1.9	31	5.4	13.44	0.72	20.3	-0.8	18.9	-0.2
15	23.9	-0.8	30.5	-0.5	12.8	-2	19.9	0.6	18.1	-3.1	30.6	-0.4	13.06	-0.38	18	-2.3	21.5	2.6
16	22.9	-1	31.3	0.8	11.4	-1.4	17.3	-2.6	14.1	-4	31.3	0.7	12.28	-0.78	14.8	-3.2	18.6	-2.9
17	21.5	-1.4	30.2	-1.1	9.7	-1.7	16.4	-0.9	11.3	-2.8	30.3	-1	10.74	-1.54	12.7	-2.1	16.1	-2.5
18	19.5	-2	24.7	-5.5	8.4	-1.3	14.9	-1.5	9.6	-1.7	24.7	-5.6	8.74	-2	9.7	-3	14.7	-1.4
19	16.4	-3.1	17.5	-7.2	6.7	-1.7	11.2	-3.7	7	-2.6	17.5	-7.2	5.5	-3.24	6.4	-3.3	11.5	-3.2
20	12.1	-4.3	11.8	-5.7	5.4	-1.3	7.7	-3.5	5	-2	11.8	-5.7	3.61	-1.89	5.1	-1.3	9.1	-2.4
21	9.4	-2.7	11.1	-0.7	5.8	0.4	7	-0.7	3.8	-1.2	11.1	-0.7	4.27	0.66	4.6	-0.5	7.2	-1.9
22	7.9	-1.5	11.2	0.1	6.4	0.6	6.3	-0.7	3.5	-0.3	11.2	0.1	5.17	0.9	4.4	-0.2	6.5	-0.7
23	7.3	-0.6	11	-0.2	7	0.6	6.4	0.1	3.1	-0.4	11	-0.2	5.65	0.48	4.3	-0.1	6.4	-0.1
24	6.7	-0.6	10.2	-0.8	8.1	1.1	6.4	0	2.8	-0.3	10.2	-0.8	6.03	0.38	4.1	-0.2	6.3	-0.1

$a = j_{NO_2}/k_3$, $b =$ differences.

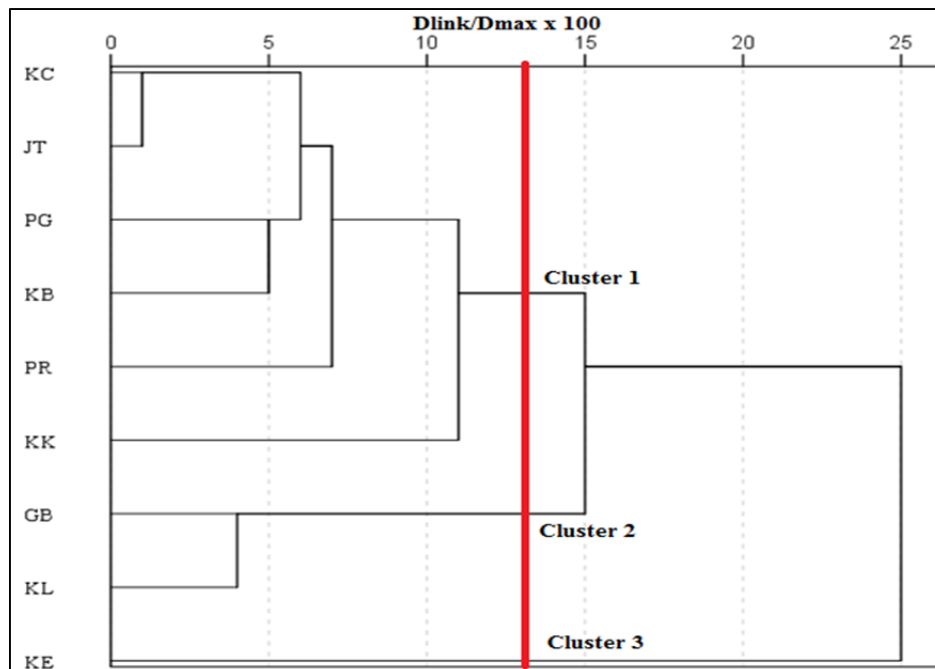


Fig. 6. Dendrogram of cluster analysis for ground level ozone.

Cluster 3 accommodated only on KE station. The differences between O_3 concentrations in KE station and other locations are believed to contribute to the results. O_3 in Kemaman was high (15.6 ppb), whereas O_3 levels were relatively low

in the two cluster 10.6 and 11.0 ppb for cluster 1 and cluster 2, respectively.

The stations in Cluster 1 are in the main city centres of each state. These stations are surrounded by high-populated

residential areas, major roads and commercial areas such as KK and KB stations. Furthermore, part of monitoring stations in cluster 1 such as KC, PG and PR stations are located in industrial areas and have the most famous ports in Malaysia namely Penang port and Port of Pasir Gudang. As example, Penang port handled a total of 6650 arrivals and departures of vessels in 2012, which is equivalent to 43 million tons of cargo and can be considered as possible sources of ozone precursors. The location of the monitoring stations is also influenced by industrial, traffic emission and power plants especially in PR and KC stations. Cluster 2 consists of stations located in the Klang Valley. In these areas, there is a very high concentration of commercial and industrial activities with heavy traffic almost entirely around the clock. Cluster 3 (Kemaman) is with a total area of 2535 km² and total population 166,750 in 2010. Only two major industrial sites (i.e., Kerteh Petrochemical and Gebeng Industrial Area) are located in and near Kemaman (Ismail *et al.*, 2011). However, the higher ozone level in this location comparing to others may be due to transport of air pollutants from other locations and several meteorological parameters which always been associated with ozone variations such as wind speed and directions. Wind speed and direction are significant agent that controlling ozone transport and dilution in both daytime and night time (Ghazali *et al.*, 2010; Kim and Guldmann, 2011; Toh *et al.*, 2013). Awang *et al.* (2015) reported that high O₃ concentrations in Kemaman were observed that coincided with prevailing winds from the southerly direction. Kuantan is one of the biggest cities in east coast Peninsular Malaysia and characterized by high population and traffic density, numbers of industrials and residential establishments.

PCA Analysis during CCT

Principal component analysis was applied on nine significant parameters of the data set that influence the formation of CCP to determine the major sources of the variation in each cluster produced by HACA. They were SO₂, PM₁₀, CO, NO₂, NO, UVB, WS, temperature, and relative humidity for each cluster. These variables were selected based on their relationship with O₃ as NO₂, NO and CO is significant O₃ precursors, PM₁₀ and SO₂ are primary pollutant and UVB, WS, temperature and relative humidity are important meteorological parameters that contribute to O₃ fluctuations. Most of the correlations between variables are significant at 0.01 level, thus allowed these variable to be used in PCA.

In addition, KMO and Bartlett's were used to justify the hypothesis and applicability of the selected variables for PCA. KMO value for cluster 1, cluster 2 and cluster 3 are 0.834, 0.815 and 0.661, respectively which showed that the value is higher than 0.5. According to Chattopadhyay and Chattopadhyay (2011), KMO could give an indication whether PCA is suitable for removing multicollinearity in the dataset and KMO value closer to 1 indicates that the correlation pattern is relatively compact and suitable for PCA. In the meantime, Sousa *et al.* (2007) reported that the null hypothesis of PCA is the variables are uncorrelated and to justify the hypothesis and applicability of the monitoring records for PCA, Bartlett's test was used. Result of the Bartlett's tests is significant as the p-value of the test is higher than 0.001 that indicate that the selected variable were related to one another and suitable for PCA.

The results of the PCA loadings after rotation are shown in Table 4. A spatial variation was observed in the loaded component of each PC and in the total variance explained by each PCA. In this study, only strong factor loadings (> 0.40) were selected for the PC interpretation.

a) Cluster 1

Cluster 1 consists of six stations that located in urban and industrial locations. The first component (PC1) explains 51.6% of the total variance which shows strong positive factor loadings for humidity (0.941), CO (0.794), NO (0.729), strong negative loadings on ambient temperature (-0.922), UVB (-0.912) and WS (-0.767). Several studies of O₃ in urban areas have shown that the variation of O₃ be influenced by a number of O₃ precursors that predominantly originate from motor vehicles such as NO and CO (Ma *et al.*, 2012; Zhang *et al.*, 2013). As stated previously in Eq. (3), in the presence of both high temperatures and sunlight NO will influence O₃ formation through titration reaction (Ghazali *et al.*, 2010). Humidity is another factor that influences formation of CCP of O₃. High RH condition enhanced O₃ destruction as a result of the reduction in photochemical efficiency and the increase in wet deposition process (Kovac-Andric *et al.*, 2009; Toh *et al.*, 2013).

The second component, (PC2) explains 19.24% of total variance. It has strong positive loadings on PM₁₀ (0.758), NO₂ (0.712) and SO₂ (0.683). Cluster 1 is a combination of urban and industrial cities; thus the burning of fuels in automobiles and industrial facilities are suspected to be the source of air pollution (SO₂, NO₂) as indicated in previous studies (Janssen *et al.*, 2001; Dominick *et al.*, 2012).

Table 4. Principal Components Analysis after varimax rotation.

Group	KMO	Cumulative Variance (%)	PC	Eigenvalue	Variance (%)	Factor loadings								
						NO ₂	NO	SO ₂	CO	PM ₁₀	T	RH	WS	UVB
Cluster 1	0.834	70.84	PC1	4.644	51.60		0.729	0.794			-0.922	0.941	-0.767	-0.912
			PC2	1.731	19.24	0.712	0.683	0.758						
Cluster 2	0.815	70.00	PC1	4.372	48.58		0.720				-0.927	0.936	-0.582	-0.892
			PC2	1.927	21.41	0.784	0.743	0.654	0.778					
Cluster 3	0.661	68.00	PC1	3.005	33.39						0.888		0.792	0.853
			PC2	1.747	19.41		0.684	0.866						
			PC3	1.368	15.20	0.758	0.697	0.603		0.715				

Meanwhile, PM_{10} is one of the most notable criteria pollutants because it can alter the photolysis rates of several trace gases. Bian and Zender (2003) claimed that high PM_{10} levels in ambient air can trigger light scattering of solar radiations and reduce the solar radiation intensity that reached ground level. Reduction in solar intensity stopped photochemical reactions and diminished O_3 concentrations.

b) Cluster 2

Cluster 2 consists of two stations GB (Gombak) and KL (Klang). The first component (PC1) explains 48.58% of the total variance. It recorded strong positive factor loadings for humidity (0.936), NO (0.720), strong negative loadings on ambient temperature (−0.927), UVB (−0.892) and WS (−0.582). As discussed in Cluster 1 motor vehicles emissions are the major source of NO and the influence of meteorological parameters is clear on CCP formation but lower loading in cluster 2. The second component (PC2) with 21.41% of total variance shows strong positive loadings on NO_2 (0.784), PM_{10} (0.778), SO_2 (0.743) and CO (0.654). The results show that the sources of pollutants in this cluster mostly related to motor vehicles and industrial emission in the area. The air monitoring stations (GB and KL) are located in dense-development urban areas in the Klang Valley and are served by several major highways, which experience heavy traffic in the morning and late afternoon rush hours. In addition, the KL station is also surrounded by industrial areas and a proximity to busy port (Port Klang) all of which have the capability to emit high amounts of pollutants into the atmosphere. According to Haris and Aris (2013), Port Klang is the busiest port in Malaysia and the 14th busiest port in the world. In 2012, the port received and departed approximately 15,000 vessels, handling approximately 169 million tons of cargo, which is significantly higher than that of any other port in Malaysia.

c) Cluster 3

In the case of Cluster 3, there is only one station (KE station). The first component (PC1) explains 33.39% of the total variance and has strong positive loadings on ambient temperature (0.888), UVB (0.853) and WS (0.792). The second component (PC2) explains 19.4% of the total variance and shows strong positive loadings for CO (0.866) and NO (0.684), which are obviously indicative of emissions from vehicles engines such as cars and lorries. The emissions of CO arises during the incomplete combustion of fossil fuels and biomass in fumes produced by portable generators and vehicle engines (USEPA, 2013). The third first component (PC3) explains 15.20% of total variance which shows strong positive factor loadings on NO_2 (0.758), RH (0.715) and PM_{10} (0.603). PM_{10} and NO_2 can be formed through open burning or emitted by motor vehicles or result from transboundary pollutants around the study area (Md Yusof *et al.*, 2010).

By comparing the results of PCAs from the three clusters, the PCs in the three clusters explained between 68% and 70.8% of the variation in ozone during CCT. Furthermore, differences between PCs in clusters 1, 2 and 3 can be observed by examining how the pollutants and

meteorological variables were loaded. Meteorological parameters have a greater influence on O_3 formation in cluster 1 (51.6%) and 2 (48.5%) than in cluster 3 (33.3%). Further, pollutant concentration parameters have a greater influence on O_3 formation differently in the three clusters in descending order 21.4%, 19.2%, 15.2% for clusters 2, 1 and 3, respectively. This difference is due to variation in the numbers of vehicles, industrial zones and number of population in each cluster. Therefore, the wise to apply a targeted spatial control strategy for ground-level ozone control during the CCT period from 8:00 to 11:00 a.m. to reduce the O_3 precursor pollutants.

CONCLUSIONS

This study analyzed the spatial variability of CCP for O_3 concentrations in nine monitoring stations located in Malaysia from 1999 to 2010. The study showed that high variability in CCP was observed between the monitoring stations which ranged from 8 a.m. and 11 a.m. The earliest time were measured at Kota Kinabalu due to the earlier sunrise at the location. At most of the industrial stations, the CCP were measured around 9 a.m.–10 a.m. and at most of the urban station CCP were measured around 10 a.m.–11 a.m. The results from this study show that the HACA grouped the nine air monitoring stations into three different clusters based on the selected parameters that influence CCP formation. Results of PCA for the three clusters showed that the contributions to O_3 level variation during CCT by meteorological variables (UVB, temperature, relative humidity, and wind speed) are higher at 51.6%, 48.5%, and 33.3% than that of primary air pollutants (NO_2 , SO_2 , PM_{10}) at 19.2%, 21.4%, and 15.2% for cluster 1, cluster 2, and cluster 3, respectively. The main sources of O_3 precursors that contribute in CCP in Cluster 1 and Cluster 3 are motor vehicle exhaust emissions and gases released from industrial activities. Meanwhile, the main sources of CCP of ground level ozone in Cluster 2 are predominantly from motor vehicles, emissions from industries and port related activities. In recommendation, all relevant agencies could collaborate to implement a State Implementation Plan (SIP) by location and by time which could lead to minimize in O_3 precursors' emission reductions such as motor vehicle exhaust emissions and well as in gas emissions from industry sectors.

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