Ground Level Ozone Fluctuational Characteristics within Two Industrial Areas in Malaysia

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Abstract. This study aims to investigate the fluctuation characteristics and source signature of ground level ozone (GLO) at a receptor site in commercial urban-industrial and suburban industrial area in Shah Alam, Selangor, and Bakar Arang are hot spots for industrialization. The fluctuation characteristics of ozone in industrial is determined using critical conversion time (CCT) and introduce the Critical Transformation Time (CTT) using secondary data from 2000 to 2011. We also use Principal Component Analysis (PCA) to determine the primary sources from atmospheric parameter and meteorological parameter. It was observed that suburban Bakar Arang has earlier CCT between 8.00 a.m. and 9.00 a.m. compared to Shah Alam where CCT occurred between 10.00 to 11.00 a.m. Results of PCA indicate ozone CCT fluctuation contribution in Shah Alam by primary air pollutants (CO, NO, NO₂, CH₄) and meteorological influence are 64.1% and 41.5%, respectively, higher than that in Bakar Arang. The use of CCT and CTT show specific time range of ozone production and destruction. These sources of compounds lead to formation of GLO and affecting the CCT. The application of CTT could be a signal of the urgent need to manage nitrogen emission from commercial industrial areas.

1. Introduction

As a developing country, Malaysia has experienced large scale land use change with increasing urbanization and industrialization [1] that lead to significant ozone pollution. Ground-level ozone (O₃) pollution is the second most significant prominent air pollution in Malaysia [2].

Ozone becomes harmful at ground level as its unstable and reactive oxidant that can deposit onto most surfaces, including biological tissues, for example, lungs, eyes or plants [3,4]. Ozone is produced by its precursors; both nitric oxide (NO) and nitrogen dioxide (NO₂) are directly involved in O₃ photochemical reactions to produce O₃. The lifetime of these precursors, the rates of their reaction in the atmosphere and the lifetime of O₃ in the troposphere combine to make tropospheric O₃ a regional-global scale problem [5]. For example, in most urban environment, such as the Klang Valley,

The diurnal cycles of surface ozone have a uni-modal shape, which is controlled by various processes, including photochemistry, physical/chemical removal, and the rate of deposition and transport [6].

In Malaysia, a number of previous studies have reported understanding the critical transformation characteristics of O₃ using NO₂ photolysis rate [6]. The critical conversion point (CCP) could be utilised to explain the various fluctuation characteristics of O₃ and predict their diurnal variations. Literature has emerged that shows the entire process of photochemical reactions which involve NO₂
photolysis and NO titration reactions where it can determine the critical conversion point (CCP) and critical conversion time (CCT) of precursors to O₃ could be deduced from them [6,7]. The findings of CCT make an important contribution to the exact time where the O₃ transformation is at its highest rate. The identification of Critical Transformation Time (CTT) in this study were set out to focus on the time range of O₃ transformational behavior before the O₃ reached it peaks level.

In this study, the CTT ozone fluctuation characteristics in two industrial areas with different background land use is presented to focus on the time range of O₃ transformational behavior before the O₃ reached it peaks level and highlight the challenges involved in ozone mitigation.

2. Methodology

2.1 Study Area

The study areas chosen for this are in Shah Alam (SA), Selangor, and Bakar Arang (BA), Kedah, where continuous air quality monitoring stations (CAQMS) are located. The CAQMS in these two locations are managed by a private company, Alam Sekitar Malaysia Sdn Bhd (ASMA) hired by Department of Environment (DOE) Malaysia. Figure 1 and 2 show the map of the location for CAQMS.

2.1.1 Shah Alam, Selangor.

Shah Alam (3.0733° N, 101.5185° E) is an established urban area in Selangor. It is the state capital of Selangor and a well-known industrial city in Malaysia with large coverage of industrial and transportation land uses [1]. The sampling station is located within 10 meters from the CAQMS located at Taman Tun Dr. Ismail residential area, connected by six major highways. Shah Alam is a heavily industrialized area with high human population and high traffic density, together with high pollutant emission. The temperature in Shah Alam varies between 33 °C and 27 °C throughout the year. March is the warmest month of the year at Shah Alam with little precipitation and November is the wettest month due to the northeast monsoon season. Shah Alam was chosen for analysis due to its high population density with urban-industrial environment hence an adverse affecting the health.

2.1.2 Bakar Arang, Sungai Petani, Kedah.

Bakar Arang, Sungai Petani, Kedah is located in Northern Peninsular Malaysia with 421,530 people. It is located on outskirts of Sungai Petani and 30 km from Penang Port. The CAQMS located at Sek. Keb. Bakar Arang near to industrial areas focused on the heavy industries from timber products, health products, electrical and electronics, plastics, textile, automotive, motor assemblies, pharmaceuticals and concrete products [8]. This area experienced uniformly high temperature in between 27°C to 30°C with mean rainfall of 267 cm. Population density and traffic density had increased therefore is expected that O₃ concentration in the sampling site will be high compared to other area. Awang et al. [9] reported that suburban Bakar Arang mean O₃ concentration is higher compared to urban mean O₃ concentration in Seberang Perai (industrial) and Kajang (urban). In conclusion, the mean O₃ concentration of Bakar Arang affected from vehicular emissions [9] and this study has focused on time range of O₃ accumulation associated with the rate of NO/NO₂ that has its time range variation in different land use environment.
Figure 1. Location of monitoring site and Continuous Air Quality Monitoring Station (CAQMS) in Shah Alam

Figure 2. Location of monitoring site and Continuous Air Quality Monitoring Station (CAQMS) in Bakar Arang
2.2 Secondary Data Collection

Twelve years of continuous hourly $O_3$ concentrations and other air pollutant levels starting from 1st January 2000 to 31st December 2011, was obtained from Department of Environment, Malaysia. There were two categories for data collection. First was to determine the fluctuation behavior $O_3$ concentration at the 2 CAQMS and identification of CTT of $O_3$ concentration using composite diurnal plot of NO-NO$\textsubscript{2}$-$O_3$. Second was to determine the sources contribute the $O_3$ concentration using Principal Component Analysis (PCA) with 11 other variables were initially arrange by month and year based on CTT. The 11 variables studied were divide into two latent factors; air pollutants and meteorological parameters.

The first latent factor is air pollutants consisted of ground level ozone ($O_3$), carbon monoxide (CO), nitrogen oxide (NO), nitrogen dioxide (NO$\textsubscript{2}$), sulphur dioxide (SO$\textsubscript{2}$), the organic pollutant group consisted of methane (CH$\textsubscript{4}$) and non-methane hydrocarbon (NMHC) while the second latent factor is meteorological parameters consisted of wind speed, ambient temperature, relative humidity and ultraviolet-B radiation (UVB). No imputation procedure was adopted for the treatment of missing data.

2.3 Primary Data Collection

This study used primary data that was collected to verify the secondary data that used to obtain the CCT ($O_3$, NO and NO$\textsubscript{2}$). The monitoring set up is divided into three categories which the location for monitoring, the period and the monitoring equipment both for air pollutants and meteorological parameters.

The important to do the primary data collection was to verify the secondary data to determine whether the secondary data is following the current trend of air quality. The location for monitoring site is located less than 1 km from CAQMS for SA and BA.

2.3.1 Monitoring Period and Equipment.

The monitoring period was started from 12.00 a.m. to 12.00 a.m. (24 hours) for 72 hours in Shah Alam and Bakar Arang. The primary data that were obtained at the studied site consists of ozone ($O_3$) and nitrogen dioxide (NO$\textsubscript{2}$). They were monitored and collected by using the specific equipment (Table 1) which was run directly on the site.

Table 1. Specific equipment used for primary data’s collection

| Type of equipment                          | Parameter                                      |
|-------------------------------------------|------------------------------------------------|
| Aeroqual S 500 Series                     | Ozone ($O_3$) and Nitrogen Oxides (NO, NO$\textsubscript{2}$) |
| Automatic Weather Station (AWS) – RK900-01| Temperature, wind speed, relative humidity      |

The Aeroqual S 500 series typically used for short term air quality assessment and monitoring. It has an interchangeable sensor that attached to the monitor base. It consists of different sensor codes, types, detection range and has the minimum limit for the detection range. Aeroqual S 500 yielded close agreement with hourly average observations from a reference instrument used by DOE in temperate ambient conditions [10,11]. The concentration of $O_3$ and NO$\textsubscript{2}$ were measured by using the same device with different portable sensor head. In term of quality control and quality assurances, Aeroqual provided a Calibration Accessory (R42) to enhance humidity control and delivery of calibration gas to the sensor head. Zero calibration is available whenever the sensors exhibited baseline reading higher than zero.

Automatic Weather Station RK900-01 is used for primary data collection for wind speed sensor with measurement accuracy at $\pm (0.3\pm0.3V)$ m/s. Wind direction sensor with measurement accuracy at $\pm3^\circ$, temperature with accuracy at $\pm5^\circ$C and relative humidity with accuracy level at $\pm3\%$, respectively.
2.4 Diurnal Variations
This study has used composite diurnal plots of \( \text{O}_3 \), NO and \( \text{NO}_2 \). Diurnal plot is representing the variation of ozone transformational behaviour together with its precursors- NO and \( \text{NO}_2 \) during daytime and night time (24 hours).

CCP can be deduced from composite diurnal plot and showed \( \text{O}_3 \) production point that has introduced as critical conversion point (CCP). CCP was occurred at a specific time and has been introduced as critical conversion time (CCT) [6]. In addition, \( \text{O}_3 \) accumulation has the time range and could be different in different environment settings which has been introduced in this study as critical transformation time (CTT) and representing the novelty of this work.

The composite diurnal plot plays an important role in this study to determine at which point the three variables (\( \text{O}_3 \), NO and \( \text{NO}_2 \)) intercept and causes the critical conversion point (CCP) to occur at specific time (CCT). From there, the critical transformation time (CTT) has been investigated. Previous research has only focused on time of \( \text{NO}_2 \) photolysis starts to accumulate (CCP). Through the use of CTT, the \( \text{NO}_2 \) photochemical reaction would be able to capture the \( \text{O}_3 \) accumulations before the \( \text{O}_3 \) decrease entering the phase of NO titration taken place. The time range of CTT could be highlighted to reduce the sources on the time range and could mitigate the exceedances of ozone concentration.

2.5 Principal Component Analysis (PCA)
PCA is widely used to reduce variables and to identify the relevant variables as sources of \( \text{O}_3 \) formation. It has capability to detect most significant variables in dataset with minimum loss of original information [6].

Equation 1 of Principal Components (PCs) as follow:

\[
P_{Ci} = l_{1i}X_1 + l_{2i}X_2 + \ldots + l_{mi}X_m
\]

In this Equation 1, where \( P_{Ci} \) is the \( i^{th} \) PC, and \( l_{mi} \) is the loading of the observed variable \( X_m \).

The first PCs is the linear combination of X having the largest variance [12]. The new variables known as PCs is defined by the eigenvectors od the covariance matrix of X. Ul-Saufie et al. [13] explained the eigenvalues shows the eigenvectors and PCs; for each PC, only eigen values larger or equal to 1 are considered as significant. Varimax rotation for PCA is important to ensure each variable is significant and correlate with only one component and has minimal association with other components. The load for each variable has significant where the factor loading is considered strong with the value 0.5, moderate with 0.4 and weak with the value 0.3.

3. Results and Discussion
The fluctuation of ozone concentration and its precursors during daytime and night time are linked to changes of critical conversion point (CCP), critical conversion time (CCT) and critical transformation time (CTT) in transformational behaviour of ozone concentration for SA and BA.

3.1 Diurnal Variations: Critical Conversion Point (CCP) and Critical Conversion Time.
The determination of CCP and CCT is analysed using composite diurnal plot from monitoring site is presented in Figure 3. \( \text{O}_3 \) diurnal variation shows the uni-modal shape at both locations. The \( \text{O}_3 \) concentration increased after sunrise, reaching a peak approximately between 12.00 p.m. and 4.00 p.m. \( \text{O}_3 \) production is characterized by photochemical reaction of \( \text{NO} \), linked to UV \(_{B}\) intensity [14].

Both SA and BA daily average concentration during monitoring do not exceed 100 part per billion (ppb) of Recommended Malaysian Ambient Air Quality Guideline (RMAAQG). The result of SA ozone started to rise around 10.00 a.m and the interception of ozone and nitrogen dioxide is obtained around 11.00 a.m to 12.00 p.m. The CCP is showing the time and known as CCT. While in BA, ozone started to rise up at 9.00 a.m. and CCT slightly earlier approximately between 8.00 a.m. and 9.00 a.m.
However, the formation fluctuation of ozone before reaching its peak has a certain time range. The time range can be obtained using the 12 years of secondary data and we represented the CCT and CTT as shown in Figure 4.

![Figure 4](image)

**Figure 4.** Diurnal plot giving the information of fluctuation of O₃. In Figure 4 (A and b) show the diurnal variation of the hourly average of O₃ and its precursors (NO and NO₂) at continuous air quality monitoring station (CAQMS). The fluctuational behaviour of individual illustrated using composite diurnal plot. O₃ production is characterized by photochemical processes. Overall O₃ diurnal variations show the uni-modal shape, mark the same pattern of primary data collection from monitoring days conducted as in Figure 3.

Based on previous studies where the researcher used diurnal plot to determine CCP of selected location [6]. Figure 4 (A and B) represent the industrial urban and sub urban CAQMS. Generally, at the beginning of the plot, NO₂ concentration was high while O₃ concentration remained low due to NOₓ titration during night time [15]. Changes started to occur when entering CCT, starting around 8.00 a.m., due to the increasing solar radiation that triggers photochemical reactions. During CCT, NO₂ was used up for the O₃ formation. CCP occurred when photochemical reaction rate started to surpass NOₓ titration rate and it was represented by the interception point between O₃ and NO₂ concentration on the plot [6, 7, 15, 16,17].

### 3.2 Critical Transformation Time (CTT)

Diurnal plot giving the information of fluctuation of O₃. In Figure 4 (A and b) show the diurnal variation of the hourly average of O₃ and its precursors (NO and NO₂) at continuous air quality monitoring station (CAQMS). The fluctuational behaviour of individual illustrated using composite diurnal plot. O₃ production is characterized by photochemical processes. Overall O₃ diurnal variations show the uni-modal shape, mark the same pattern of primary data collection from monitoring days conducted as in Figure 3.

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The O$_3$ uni-modal peaks usually after CCT approximately between 12.00 p.m. to 4.00 p.m. For both areas, the ozone concentration starts to decrease after the CCT time (the uni-modal peaks) around 2.00 p.m. to 5.00 p.m. The decrease of O$_3$ is due to the decrease of UV intensity as the sun radiation starts to decline. As the ozone decrease, NO$_2$ starts to increase in Equation (2), known as titration process.

Ozone starts to decrease started at 7.00 p.m. to 7.00 a.m. following reactions in equations (2) – (7):

\[
\text{NO}_2 + \text{hv(}\lambda < 420\text{nm}) \rightarrow \text{NO} + \text{O}
\]  

(2)

\[
\text{O} + \text{O}_2 + \text{M} \rightarrow \text{O}_3 + \text{M}
\]  

(3)

\[
\text{NO} + \text{O}_2 \rightarrow \text{NO}_2 + \text{O}_2
\]  

(4)

\[
\text{NO}_2 + \text{O}_3 \rightarrow \text{NO}_3 + \text{O}_2
\]  

(5)

\[
\text{NO}_3 + \text{NO}_2 + \text{M} \leftrightarrow \text{N}_2\text{O}_5 + \text{M}
\]  

(6)

\[
\text{N}_2\text{O}_5 + \text{H}_2\text{O} \rightarrow 2\text{HNO}_3
\]  

(7)

During night time, the reaction between O$_3$ and NO$_2$ yield nitrate (NO$_3$) radicals through Equation (5). The produced NO$_3$ radicals directly react with NO$_2$, producing dinitrogen pentoxide (N$_2$O$_5$). It is unstable and will dissociated back to NO$_2$ and NO$_3$ radicals Equation (6). However, N$_2$O$_5$ may also undergo reaction with H$_2$O to form nitric acid (HNO$_3$). Increasing water vapour in night time relatively increase the RH will enhance the formation of nitric acid, HNO$_3$ Equation (7). HNO$_3$ is highly soluble in water and deposit thru precipitation as acid rain [3,18]. Limited night time NO$_2$ directly reduce the reaction rates of Equation (5) and Equation (6).

The decline continues until it reaches a stable concentration throughout the nights starting at 11.00 p.m. The interruption of removal reactions allowed O$_3$ to remain in the atmosphere [15].

**Figure 4.** Composite diurnal plot of O$_3$, NO and NO$_2$ concentration at industrial urban CAQMS: (A) Shah Alam and industrial suburban (B) Bakar Arang in 12 years (2000-2011)
The results showed that the CCT in Table 4.2 of each location occurred within the CTT, which was between 8.00 a.m. and 11.00 a.m. CCT shown a time where a point that has intersection point of $O_3$ and NO$_2$ concentration. From the result, it was observed that the CCT at Kota Bharu occurred at ± 8.00 a.m. while CCT at Shah Alam, Kajang and Johor Bahru occurred at ± 10.00 a.m. These four locations are in the same categories of land use however the range of CCT in Kota Bharu slightly earlier while the other locations were in the same range. This finding confirming that the photochemical reaction depending on its sources and the temperature [19] which influencing the CCT to occurred early or later in different environment settings.

Table 2. Critical conversion time (CCT) and critical transformation time (CTT) based on composite diurnal plots at SA and BA.

| Year | Critical Conversion Time (CCT) (Industrial-Urban) | Critical Conversion Time (CCT) (Industrial-Suburban) |
|------|-----------------------------------------------|---------------------------------------------------|
|      | Shah Alam                                    | Bakar Arang                                       |
| 2000 | 10.45                                         | 9.15                                              |
| 2001 | 10.55                                         | 9.30                                              |
| 2002 | 10.55                                         | 9.45                                              |
| 2003 | 10.55                                         | 9.45                                              |
| 2004 | 10.55                                         | 9.15                                              |
| 2005 | 10.45                                         | 9.10                                              |
| 2006 | 10.15                                         | 9.40                                              |
| 2007 | 10.15                                         | 9.30                                              |
| 2008 | 10.45                                         | 8.55                                              |
| 2009 | 10.30                                         | 9.05                                              |
| 2010 | 10.30                                         | 9.00                                              |
| 2011 | 10.30                                         | 9.15                                              |

Critical Transformation Time (CTT) 10.00 a.m to 11.00 a.m 8.00 a.m to 10.00 a.m

3.3 Principal Component Analysis (PCA)

The PC's results in Table 3 show after varimax rotation. Varimax rotation in PCA is use to maximize correlation of a variable with one component only and minimize of the variables with the other component [19].

Table 3. Total Variance explained of diurnal ozone

| Location | Component | Diurnal Ozone | Rotated sum of square loading | V (%) | C(%) |
|----------|-----------|---------------|-------------------------------|-------|------|
| Shah Alam| 1         |               | 7.195                         | 64.10 | 85.43|
|          | 2         |               | 1.348                         | 12.26 |      |
| Bakar Arang| 1       |               | 4.565                         | 41.498| 86.12|
|          | 2         |               | 3.790                         | 34.454|      |
|          | 3         |               | 1.118                         | 10.165|      |

Whereas, the factor loadings for each variable in Table 4 explained the ten significant parameters of the data set that influence the ozone fluctuation in SA and BA, respectively. Shah Alam diurnal ozone fluctuation is contributed by the first component with the biggest factor loading 7.195 with 64.1% of total variance. There are 8 variables listed as significant variables (e.g. NO$_2$, NO, CO, PM$_{10}$, CH$_4$, Temperature, Ultraviolet-B, Relative Humidity, Wind Speed). One variable has listed in second component (e.g. SO$_2$) with only 12.26% of total variance.
Table 4. Rotated component matrix using varimax rotation

| Location     | Shah Alam | Bakar Arang |
|--------------|-----------|-------------|
| PC           | PC1       | PC2         | PC1   | PC2   | PC3   |
| NO₂          | -0.889    | 0.884       |       |       |       |
| NO           | 0.926     | 0.904       |       |       |       |
| CO           | 0.953     | 0.909       |       |       |       |
| PM₁₀         | 0.923     | 0.835       |       |       |       |
| SO₂          |           | 0.952       | 0.956 |       |       |
| CH₄          | 0.845     | -0.812      |       |       |       |
| Temp         | -0.926    | 0.807       |       |       | 0.611 |
| UVB          | -0.806    |             |       |       |       |
| RH           | 0.885     | -0.859      |       |       |       |
| WS           | -0.883    | 0.758       |       |       |       |

Meanwhile in Bakar Arang, there are three PCs. The first factor loading is 4.565 representing 41.498% of the total variance. The first component listed are mixing of primary pollutants and meteorology parameters (NO₂, CH₄, Temperature, Relative humidity and Wind speed). PCs 2 comprise of other primary pollutants such NO, CO, PM₁₀ and SO₂. The last variable into PCs 3 is only UVB with 10.165 total variance.

In conclusion of the results presented, the commercial industrial area in SA emits more oxides of nitrogen (NO and NO₂) than in BA. These two main precursors have contributed to the time of ozone formation, the CCT and CTT. During CCT, NO₂ was used up for the O₃ formation. CCP occurred when photochemical reaction rate started to surpass NO₃ titration rate and it was represented by the interception point between O₃ and NO₂ concentration on the plot [6,7,15,16,17]. Bakar Arang CCT and CTT slightly earlier while the other locations were in the same range. This finding confirming that the photochemical reaction depending on its sources and the temperature [18] which influencing the CCT to occurred early or later in different environment settings. CCT indicated the highest rate of photochemical reaction at that point. The diurnal plot showed that the NO₂ concentration decreased after CCT while O₃ concentration increased. This indicated that the transformation of NO₂ to O₃ occurred rapidly, which lead to accumulation of O₃ and concentration of O₃ increased. The O₃ maxima happened after CTT, usually between 12.00 p.m. and 4.00 p.m. After O₃ maxima, O₃ concentration tends to decrease due to the decrease of solar energy. NOₓ titration takes place after sunset and causes NO₂ concentration to rise back and surpassed O₃ concentration.

4. Conclusion
O₃ concentration starts to increase after the sunrise, where a point for O₃ starts to produce has been introduce as CCP and CCT. The time range for O₃ to accumulate and increase had led in this study and known as CTT and being used to predict O₃ diurnal variation. The research has suggested factors need to be considered such the distribution of NOₓ with meteorological factors need to be addressed between the CAQMS. Thus, the importance of critical transformation time (CTT) could be highlight to explore the ground level ozone transformational behaviour for urban, industrial and suburban. In addition, the used of CTT in prediction of ozone variation could provide as an early signal of the urgent need to manage nitrogen emission from commercial-industrial areas that has detrimental effects on air quality and climate change and that has to be avoided.

Acknowledgements
We would like to thank Universiti Malaysia Kelantan in support of this study through the Skim Geran Penyelidikan Jangka Pendek – SGJP (R/SGJP/A08.00/00874A/ 001/2018/000497) and Fundamental Research Grant Scheme (FRGS) (R/FRGS/A0800/01525A/003/00554).
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