



**Laporan Akhir Projek Penyelidikan Jangka Pendek**

**Modeling The Fluctuational And  
Transformation Characteristics Of Peak  
Ground Level Ozone In Malaysian  
Ambient Air**

**By**

**Prof. Dr. Nor Azam Ramli**

**Ahmad Shukri Yahaya**

**Nor Faizah Fitri Md Yusof**

**Nurul Izma Mohamed**

**2016**

**LAPORAN AKHIR**

**'THE FLUCTUATIONAL AND TRANSFORMATION  
CHARACTERISTICS OF PEAK GROUND LEVEL OZONE IN  
MALAYSIAN AMBIENT AIR'  
(RUI/1001/PAWAM/811206)**

**NOR AZAM RAMLI**

**June 2016**

## RU GRANT FINAL REPORT CHECKLIST

Please use this checklist to self-assess your report before submitting to RCMO.  
Checklist should accompany the report.

NO.	ITEM	PLEASE CHECK (✓)		
		PI	JKPTJ	RCMO
1	Completed Final Report Form	✓	✓	✓
2	Project Financial Account Statement (e-Statement)	✓	✓	✓
3	Asset/Inventory Return Form (Borang Penyerahan Aset/Inventori)	✓	✓	✓
4	A copy of the publications/proceedings listed in Section D(ii) (Research Output)	✓	✓	✓
5	Comprehensive Technical Report	✓	✓	✓
6	Other supporting documents, if any      PGC	✓	✓	✓
7	Project Leader's Signature	✓	✓	✓
8	Endorsement of PTJ's Evaluation Committee	✓	✓	✓
9	Endorsement of Dean/ Director of PTJ's	✓	✓	✓



### BORANG PENYERAHAN ASET / INVENTORI

#### A. BUTIR PENYELIDIK

1. NAMA PENYELIDIK : NOR AZAM RAMLI.  
 2. NO STAF : AE50187  
 3. PTJ : PPKA  
 4. KOD PROJEK : RUI – 1001/PAWAM/811206  
 5. TARIKH TAMAT PENYELIDIKAN : 14 JANUARI 2016.

#### B. MAKLUMAT ASET / INVENTORI

BIL	KETERANGAN ASET	NO HARTA	NO. SIRI	HARGA (RM)
1	Pemproses Data	5263	2QMYWX1	7990
2	AEROQUAL SSL 550	5347	160620142312	3868
3	AEROQUAL SSL 550	5348	160620142307	3868
4	IDRISI SELVA	5293	5293	1362.45

#### C. PERAKUAN PENYERAHAN

Saya dengan ini menyerahkan aset/ inventori seperti butiran B di atas kepada pihak Universiti:

.....  
 (  )

Tarikh:

#### D. PERAKUAN PENERIMAAN

Saya telah memeriksa dan menyemak setiap alatan dan didapati :

- Lengkap  
 Rosak  
 Hilang : Nyatakan.....  
 Lain-lain : Nyatakan .....

Diperakukan Oleh :

.....  
 Tandatangan

Pegawai Aset PTJ

Nama

Tarikh

ZULKIFLY BIN HASHIM  
 Penerimaan Jurutera  
 Pusat Pengajian Kejuruteraan Awam  
 Kampus Kejuruteraan  
 Universiti Sains Malaysia  
 Nibong Tebal, Seberang Perai Selatan,  
 Pulau Pinang  
 27/1/16

\*Nota : Sesalinan borang yang telah lengkap perlulah dikemukakan kepada Unit Pengurusan Harta, Jabatan Bendahari dan Pejabat RCMO untuk tujuan rekod.





## RU GRANT FINAL REPORT FORM

Please email a softcopy of this report to [rcmo@usm.my](mailto:rcmo@usm.my)

<b>A</b>	<b>PROJECT DETAILS</b>
<b>i</b>	<b>Title of Research: THE FLUCTUATIONAL AND TRANSFORMATION CHARACTERISTICS OF PEAK GROUND LEVEL OZONE IN MALAYSIAN AMBIENT AIR</b>
<b>ii</b>	<b>Account Number: 1001/PAWAM/811206</b>
<b>iii</b>	<b>Name of Research Leader: NOR AZAM RAMLI</b>
<b>iv</b>	<b>Name of Co-Researcher:</b> 1. AHMAD SHUKRI YAHAYA 2. NOR FAIZAH FITRI MD YUSOF 3. NURUL IZMA MOHAMED (UTP)
<b>v</b>	<b>Duration of this research:</b> a) <b>Start Date</b> : 15 Julai 2012 b) <b>Completion Date</b> : 14 Januari 2016 c) <b>Duration</b> : 42 Bulan d) <b>Revised Date (if any)</b> : -
<b>B</b>	<b>ABSTRACT OF RESEARCH</b>
	<p><i>(An abstract of between 100 and 200 words must be prepared in Bahasa Malaysia and in English. This abstract will be included in the Report of the Research and Innovation Section at a later date as a means of presenting the project findings of the researcher/s to the University and the community at large)</i></p> <p style="text-align: center;"><b>THE FLUCTUATIONAL AND TRANSFORMATION CHARACTERISTICS OF PEAK GROUND LEVEL OZONE IN MALAYSIAN AMBIENT AIR</b></p> <p>Ground level ozone (O<sub>3</sub>) is a noxious air pollutant that imposed adverse effect to human health, crop yield and the environment. Hence, it is important to understand their fluctuation and transformation characteristic which is still regarded at its infancy stage in Malaysia to properly design abatement and control strategies. Therefore, this study aimed to investigate the characteristics of O<sub>3</sub> fluctuation and transformation from its precursors as well as to introduce the critical conversion point (CCP) of O<sub>3</sub> formation. Next hour O<sub>3</sub> prediction models during daily, daytime, nighttime and critical conversion time were also developed using multiple linear regression and principal component regression to predict O<sub>3</sub> concentrations in different group of monitoring stations. This study consists of 18 areas</p>

across Malaysia from 1999 to 2010. Although an overwhelming majority of the recorded data are below the prescribed levels in the Malaysian Ambient Air Quality Guidelines, total of 1,988 hour of exceedences have been recorded with majority contributed by Shah Alam. O<sub>3</sub> diurnal cycles suggest a unimodal peak between 12 pm to 4 pm, while minimum concentration is consistently measured on 8 am. Results of PCA showed the contributions to O<sub>3</sub> variation by NO<sub>2</sub>, NO, T and UVB is up to 80%, whereas the composite diurnal plots confirmed that variation is highly depend on NO<sub>2</sub> and NO concentrations. The CCP of O<sub>3</sub> formation were identified to occur in between 8 am to 11 am. MLR model based on cluster group during daily exhibited optimal performance in terms of coefficient of determination, prediction accuracy, and index of agreement with values of 0.9351, 0.9671 and 0.9831, respectively. Although the optimal model was during daily, the developed models using critical conversion time were more consistent with minimal differences between MLR and PCR in terms of error and accuracy. Effective ground level ozone control, need strategic, systematic and concerted efforts among environmental related agencies in Malaysia

### **CIRI TURUN NAIK DAN UBAH TAMPIL TINGKAH LAKU KRITIKAL OZON PARAS TANAH DI MALAYSIA**

Ozon paras tanah merupakan pencemar udara yang berbahaya dan berupaya mendatangkan kesan negatif terhadap kesihatan manusia, hasil tanaman dan alam sekitar. Maka, pemahaman mengenai ciri-ciri turun naik dan ubah tampil ozon adalah penting untuk merangka strategi pengurangan dan pengawalan kerana kajian berkaitan ozon di Malaysia dianggap masih pada peringkat permulaan. Oleh itu, kajian ini dijalankan bertujuan untuk mengenalpasti ciri-ciri turun naik dan ubah tampil ozon berdasarkan prapenandanya serta memperkenalkan titik perubahan kritikal dalam pembentukan ozon. Model ramalan ozon untuk jam berikutnya bagi masa harian, siang, malam dan masa perubahan kritikal juga dibentuk dengan menggunakan regrasi linear berganda (MLR) dan regrasi komponen utama (PCR) untuk meramal kepekatan ozon daripada kumpulan stesyen cerapan yang berbeza. Kajian ini merangkumi 18 kawasan di Malaysia dari tahun 1999 sehingga 2010. Walaupun majoriti data yang direkodkan adalah dibawah tahap yang disyorkan oleh Garispanduan Kualiti Udara Malaysia, sejumlah 1988 jam (exceedences) telah direkodkan yang mana kebanyakannya disumbangkan oleh Shah Alam. Kitaran diurnal ozon menunjukkan puncak unimodal diantara jam 12 tengah hari ke 4 petang, manakala kepekatan minimum yang direkodkan adalah konsisten pada jam 8 pagi. Analisis PCA menunjukkan 80% daripada kepelbagaian ozon disumbangkan oleh NO<sub>2</sub>, NO, T dan UVB manakala plot diurnal komposit mengesahkan kepelbagaian ozon sangat bergantung kepada kepekatan NO<sub>2</sub> and NO. CCP pembentukan ozon dikenalpasti berlaku diantara jam 8 pagi ke 11 pagi. Model MLR kumpulan kluster harian mempamerkan prestasi paling optima berdasarkan kepada pekali penentu, ketepatan ramalan dan indeks persetujuan dengan bacaan 0.9351, 0.9671 dan 0.9831. Walaupun, model optima adalah semasa harian, model MLR dan PCR yang dibina dengan menggunakan masa perubahan kritikal adalah lebih konsisten dengan mempamerkan perbezaan minima dari segi kesilapan dan ketepatan ramalan. Pengawalan ozon permukaan yang efektif, perlukan kerjasama erat antara semua agensi yang berkaitan dengan alam sekitar.



UNIVERSITI SAINS MALAYSIA

JABATAN BENDAHARI

PENYATA PERBELANJAAN SEHINGGA 21 MAC 2016

Projek :

No. Akaun : 1001.PAWAM.811206.

Vot	Nama Fot	Peruntukan Projek	Perbelanjaan Terkumpul Sehingga Thn Lalu	Baki Peruntukan Tahun Lalu	Peruntukan Thn Semasa	Jumlah Peruntukan Thn Semasa	Tanggungjawab Semasa	Bayaran Thn Semasa	Jumlah Belanja Thn Semasa	Baki Projek
111	GAJI	72,000.00	14,988.52	57,011.48	0.00	57,011.48	0.00	0.00	0.00	57,011.48
114	ELAUN KERJA LEBIH MASA	0.00	401.40	-401.40	0.00	-401.40	0.00	0.00	0.00	-401.40
221	PERJALANAN DAN SARA HIDUP	23,000.00	26,120.10	-3,120.10	0.00	-3,120.10	0.00	0.00	0.00	-3,120.10
223	PERHUBUNGAN DAN UTILITI	0.00	1,467.10	-1,467.10	0.00	-1,467.10	0.00	0.00	0.00	-1,467.10
224	SEWAAN	7,200.00	100.00	7,100.00	0.00	7,100.00	0.00	90.00	90.00	7,010.00
225	BAHAN MAKANAN DAN MINUMAN	0.00	109.90	-109.90	0.00	-109.90	0.00	0.00	0.00	-109.90
226	BEKALAN BAHAN MENTAH	0.00	3,017.15	-3,017.15	0.00	-3,017.15	0.00	0.00	0.00	-3,017.15
227	BEKALAN DAN BAHAN LAIN	0.00	5,779.86	-5,779.86	0.00	-5,779.86	0.00	282.87	282.87	-6,062.73
228	PENYELENGGARAN & PEMBAIKAN KECIL	0.00	16,703.44	-16,703.44	0.00	-16,703.44	0.00	0.00	0.00	-16,703.44
229	PERKHIDMATAN IKTISAS & HOSPITALITI	7,000.00	26,860.08	-19,860.08	0.00	-19,860.08	0.00	4,128.10	4,128.10	-23,988.18
335	HARTA MODAL	7,999.00	17,088.45	-9,089.45	0.00	-9,089.45	0.00	0.00	0.00	-9,089.45
552	PERBELANJAAN LAIN	0.00	0.00	0.00	0.00	0.00	0.00	16.90	16.90	-16.90
<b>Jumlah</b>		<b>117,199.00</b>	<b>112,636.00</b>	<b>4,563.00</b>	<b>0.00</b>	<b>4,563.00</b>	<b>0.00</b>	<b>4,517.87</b>	<b>4,517.87</b>	<b>45.13</b>

Penyata ini adalah cetakan komputer tiada tandatangan diperlukan

Penyata ini adalah dianggap tepat jika tiada maklumbalas dalam tempoh masa 14 hari dari tarikh penyata



**C BUDGET & EXPENDITURE**

**i**

**Total Approved Budget** : RM 117,199.00

**Yearly Budget Distributed**

Year 1 : RM 39,399  
Year 2 : RM 38,400  
Year 3 : RM 39,400

**Total Expenditure** : RM 117,153.87

**Balance** : RM 45.13

**Percentage of Amount Spent (%)** : 99.96

**# Please attach final account statement (eStatement) to indicate the project expenditure**

**ii Equipment Purchased Under Vot 35000**

No.	Name of Equipment	Amount (RM)	Location	Status
1	Pemproses Data	7990	1.19PPKA	PASSED ON TO 305/PAWAM/6013607 AND 1001/PAWAM/814278
2	AEROQUAL – HH S500L	3868	1.19PPKA	PASSED ON TO 305/PAWAM/6013607 AND 1001/PAWAM/814278
3	AEROQUAL – HH S500L	3868	1.19PPKA	PASSED ON TO 305/PAWAM/6013607 AND 1001/PAWAM/814278
4	STUDENT LICENSE IDRISI SELVA SOFTWARE	1362.45	1.19PPKA	PASSED ON TO 305/PAWAM/6013607 AND 1001/PAWAM/814278

**# Please attach the Asset/Inventory Return Form (Borang Penyerahan Aset/Inventori) – Appendix 1**

**D RESEARCH ACHIEVEMENTS**

**i Project Objectives (as stated/approved in the project proposal)**

No.	Project Objectives	Achievement
1	To determine fluctuational behaviour of ozone concentration and to investigate the time high ozone concentration occurs using time series plot	Achieved
2	To understand the influence of NOx, insolation and temperature on the formation of ozone using diurnal fluctuation plot and multivariate analysis of GLO concentrations	Achieved
3	To develop the multiple linear regression model to predict ozone concentration using several performance indicator	Achieved



	to identify the best prediction model describing ozone in Malaysian urban, suburban and rural areas.	
4	To identify the optimal next hour prediction models in describing O <sub>3</sub> fluctuation in Malaysia using several performance indicators.	Achieved
5	To propose best control framework to manage the exposure and impact of GLO to public.	Achieved
6		

<b>ii Research Output</b>		
<b>a) Publications in ISI Web of Science/Scopus</b>		
No.	Publication (authors,title,journal,year,volume,pages,etc.)	Status of Publication (published/accepted/ under review)
1	<b>Norrimi Rosaida Awang, Maher Elbayoumi, Nor Azam Ramli &amp; Ahmad Shukri Yahaya (2016).</b> Diurnal variations of ground level ozone in three port cities in Malaysia. <i>Air Quality, Atmosphere and Health</i> 6(1), 25-39.	Published
2	<b>Norrimi Rosaida Awang, Maher Elbayoumi, Nor Azam Ramli &amp; Ahmad Shukri Yahaya (2016).</b> The Influence of Spatial Variability of Critical Conversion Point (CCP) in Production of Ground Level Ozone in the Context of Tropical Climate. <i>Aerosol and Air Quality Research</i> 16(1), 153-165.	Published
3	<b>Norrimi Rosaida Awang, Nor Azam Ramli, Ahmad Shukri Yahaya &amp; Maher Elbayoumi (2015).</b> High Nighttime Ground-Level Ozone Concentrations in Kemaman: NO and NO <sub>2</sub> Concentration Attributions. <i>Aerosol and Air Quality Research</i> 15(4), 1357-1366.	Published
4	<b>Norrimi Rosaida Awang, Nor Azam Ramli, Ahmad Shukri Yahaya &amp; Maher Elbayoumi (2015).</b> Multivariate Methods to Predict Ground Level Ozone during Daytime, Nighttime, and Critical Conversion Time in Urban Areas. <i>Atmospheric Pollution Research</i> 6(5), 726-734.	Published
<b>b) Publications in Other Journals</b>		
No.	Publication (authors,title,journal,year,volume,pages,etc.)	Status of Publication (published/accepted/ under review)
	<b>Norrimi Rosaida Awang, Nor Azam Ramli &amp; Ahmad Shukri Yahaya (2015).</b> Temporal Analysis of Ozone and Nitrogen Oxides Fluctuations at Pasir Gudang, Malaysia. <i>Applied Mechanics and Materials</i> (773-774), 1237-1241.	

	<b>Online</b>	
	Norrimi Rosaida Awang, Nor Azam Ramli., Ahmad Shukri Yahaya (2013). Time Series Evaluation of Ozone Concentrations in Malaysia Based on Location of Monitoring Stations. <i>International Journal of Engineering and Technology</i> 3:3, 390-394.	

**c) Other Publications**  
(book, chapters in book, monograph, magazine, etc.)

No.	Publication (authors, title, journal, year, volume, pages, etc.)	Status of Publication (published/accepted/ under review)

**d) Conference Proceeding**

No.	Conference (conference name, date, place)	Title of Abstract/Article	Level (International/National)
1	<i>International Integrated Engineering Summit 2014</i> , Universiti Tun Hussein Onn.	Norrimi Rosaida Awang, Nor Azam Ramli, Ahmad Shukri Yahaya (2014). Temporal Analysis of Ozone and Nitrogen Oxides Fluctuations at Pasir Gudang, Malaysia.	
2	<i>Seminar Hasil Penyelidikan Sektor Pengajian Tinggi Kementerian Pendidikan 2013(SHP-KPT2013)</i> , Universiti Utara Malaysia.	Norrimi Rosaida Awang, Nor Azam Ramli, Ahmad Shukri Yahaya (2013). Comparative Studies of Ground Level Ozone Concentration in East and West Coast Peninsular Malaysia.	
3	<i>4<sup>th</sup> International Conference on Human Habitat and Environment</i> , Universiti Kebangsaan Malaysia. 2013	Norrimi Rosaida Awang, Nor Azam Ramli, Ahmad Shukri Yahaya (2013).	
4	<i>3<sup>rd</sup> International Conference</i>	Norrimi Rosaida Awang, Nor	

	<i>on Human Habitat and Environment</i> , Universiti Kebangsaan Malaysia. 2013.	Azam Ramli, Ahmad Shukri Yahaya (2012). Review of Fluctuation Ground Level Ozone Concentration and Its Precursors in Peninsular Malaysia.	

**# Please attach a full copy of the publication/proceeding listed above**

**iii Other Research Output/Impact From This Project**  
(patent, products, awards, copyright, external grant, networking, etc.)

Networking with The City University of Hong Kong. (Profesor Peter Brimblecombe, External Examiner for PhD graduated from this grant)

**E HUMAN CAPITAL DEVELOPMENT**

**a) Graduated Human Capital**

Student	Nationality (No.)		Name
	National	International	
PhD			1. Dr Norrimi Rosaida Awang 2.
MSc			1. Lisa Tan Wei Chen 2. Siti Aminah Md Saad
Undergraduate			1. 2.

**b) On-going Human Capital**

Student	Nationality (No.)		Name
	National	International	
PhD			1. Nazatul Syadia Zainordin 2. Maisarah Sulaiman
MSc			1. 2.
Undergraduate			1. 2.

**c) Others Human Capital**

Student	Nationality (No.)		Name
	National	International	
Post Doctoral Fellow		1	1. Dr Maher ElBayoumi (1 tahun) 2. Dr Norrimi RosaidaAwang (2 bulan)

Research Officer			1. 2.
Research Assistant			1. 2.
Others (.....)			1. 2.

**F COMPREHENSIVE TECHNICAL REPORT**

Applicants are required to prepare a comprehensive technical report explaining the project. The following format should be used (this report must be attached separately):  
Report is attached.

**G PROBLEMS/CONSTRAINTS/CHALLENGES IF ANY**

*(Please provide issues arising from the project and how they were resolved)*

This research is labour intensive and needs long time outdoor monitoring during certain part of the research. Hence many parttime human capital were involved and they are paid honorarium to attract their interest. Instruments broke down during research. This was solved by purchasing two new units of portable monitoring equipment. Thank to USM..

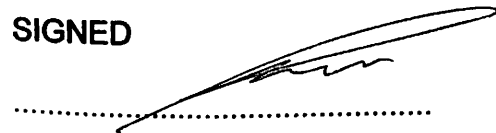
**H RECOMMENDATION**

*(Please provide recommendations that can be used to improve the delivery of information, grant management, guidelines and policy, etc.)*

HEPG made online to ensure progress from application to approval easier and kept paperless.

**Project Leader's Signature:**

SIGNED



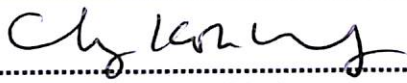
Name : NOR AZAM RAMLI

Date : 23 Jun 2016



I COMMENTS, IF ANY/ENDORSEMENT BY PTJ'S RESEARCH COMMITTEE

The research study with some as a valuable resource for peak ground level store in Malaysia. The research study has also produced impactful publication and trained human capital in terms of PhD holders and postdoctoral research.



Signature and Stamp of Chairperson of PTJ's Evaluation Committee

Name :

Assoc. Prof. Dr. Choong Kok Keong  
Deputy Dean  
Research, Postgraduate & Networking  
School of Civil Engineering  
Engineering Campus  
Universiti Sains Malaysia

Date : 27/6/2016



Signature and Stamp of Dean/ Director of PTJ

Prof. Dr. Ahmad Farhan Mohd Sadullah  
Dekan  
Pusat Pengajian Kejuruteraan Awam  
Kampus Kejuruteraan  
Universiti Sains Malaysia

Name :

Date : 27/6/16



UNIVERSITI SAINS MALAYSIA

JABATAN BENDAHARI

PENYATA PERBELANJAAN SEHINGGA 21 MAC 2016

Projek :

No. Akaun : 1001.PAWAM.811206.

Vot	Nama Fot	Peruntukan Projek	Perbelanjaan Terkumpul Sehingga Thn Lalu	Baki Peruntukan Tahun Lalu	Peruntukan Thn Semasa	Jumlah Peruntukan Thn Semasa	Tanggung Semasa	Bayaran Thn Semasa	Jum Belanja Thn Semasa	Baki Projek
111	GAJI	72,000.00	14,988.52	57,011.48	0.00	57,011.48	0.00	0.00	0.00	57,011.48
114	ELAUN KERJA LEBIH MASA	0.00	401.40	-401.40	0.00	-401.40	0.00	0.00	0.00	-401.40
221	PERJALANAN DAN SARA HIDUP	23,000.00	26,120.10	-3,120.10	0.00	-3,120.10	0.00	0.00	0.00	-3,120.10
223	PERHUBUNGAN DAN UTILITI	0.00	1,467.10	-1,467.10	0.00	-1,467.10	0.00	0.00	0.00	-1,467.10
224	SEWAAN	7,200.00	100.00	7,100.00	0.00	7,100.00	0.00	90.00	90.00	7,010.00
225	BAHAN MAKANAN DAN MINUMAN	0.00	109.90	-109.90	0.00	-109.90	0.00	0.00	0.00	-109.90
226	BEKALAN BAHAN MENTAH	0.00	3,017.15	-3,017.15	0.00	-3,017.15	0.00	0.00	0.00	-3,017.15
227	BEKALAN DAN BAHAN LAIN	0.00	5,779.86	-5,779.86	0.00	-5,779.86	0.00	282.87	282.87	-6,062.73
228	PENYELENGGARAN & PEMBAIKAN KECIL	0.00	16,703.44	-16,703.44	0.00	-16,703.44	0.00	0.00	0.00	-16,703.44
229	PERKHIDMATAN IKTISAS & HOSPITALITI	7,000.00	26,860.08	-19,860.08	0.00	-19,860.08	0.00	4,128.10	4,128.10	-23,988.18
335	HARTA MODAL	7,999.00	17,088.45	-9,089.45	0.00	-9,089.45	0.00	0.00	0.00	-9,089.45
552	PERBELANJAAN LAIN	0.00	0.00	0.00	0.00	0.00	0.00	16.90	16.90	-16.90
<b>Jumlah</b>		<b>117,199.00</b>	<b>112,636.00</b>	<b>4,563.00</b>	<b>0.00</b>	<b>4,563.00</b>	<b>0.00</b>	<b>4,517.87</b>	<b>4,517.87</b>	<b>45.13</b>

Penyata ini adalah cetakan komputer tiada tandatangan diperlukan

Penyata ini adalah dianggap tepat jika tiada maklumbalas dalam tempoh masa 14 hari dan tarikh penyata



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

Norrimi Rosaida Awang, Maher Elbayoumi\*, Nor Azam Ramli, Ahmad Shukri Yahaya

Clean Air Research Group, School of Civil Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia

### ABSTRACT

Critical conversion point (CCP) is a very crucial step in production of the ground level O<sub>3</sub> 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 O<sub>3</sub> 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<sub>2</sub>, SO<sub>2</sub>, PM<sub>10</sub>) 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 O<sub>3</sub> precursors during the CCT period is a crucial step.

**Keywords:** NO<sub>2</sub> photolysis; NO titration; Critical conversion point; Multivariate analysis.

### INTRODUCTION

Ground-level ozone (O<sub>3</sub>) 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 O<sub>3</sub> are induced by the emissions of nitrogen oxide (NO<sub>x</sub>) and volatile organic compounds (VOCs) (Seinfeld and Pandis, 2006). O<sub>3</sub> formation is very responsive to changes in meteorological parameters. Thus, elevated O<sub>3</sub> 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 O<sub>3</sub> formation toward UV light causes its clear daily variations. In the presence of sunlight, nitrogen dioxide (NO<sub>2</sub>) undergoes photochemical reactions to produce free oxygen atom (O), which later reacts with oxygen molecules (O<sub>2</sub>) to form O<sub>3</sub> (Duenas *et al.*, 2004; Azmi *et al.*, 2010). Once O<sub>3</sub> is created, it is destroyed through

several pathways, such as nitric titration and surface deposition (Abdul-Wahab *et al.*, 2005). O<sub>3</sub> concentration variations show an interesting patterns in the morning where O<sub>3</sub> level reaches the lowest concentration because of the higher rate of NO titration (Jiménez-Hornero *et al.*, 2010). Once the minimal point is reached, O<sub>3</sub> starts to increase with rising NO<sub>2</sub> concentration, thereby promoting NO<sub>2</sub> photolysis. When the NO<sub>2</sub> photolysis rate is higher than the NO titration rate, critical conversion point (CCP) occurs. Therefore, CCP is very crucial step in ground-level O<sub>3</sub> chemistry because the different in the chemical reaction's rate is expected to result in O<sub>3</sub> accumulation.

The background O<sub>3</sub> 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 O<sub>3</sub> condition and have set guidelines against this air pollutant. In 2010, the Recommended Malaysian Air Quality Guideline (RMAQG) of 100 ppbv for the hourly O<sub>3</sub> 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 O<sub>3</sub>, its precursors, and meteorological parameters occur within a wide range

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# Diurnal variations of ground-level ozone in three port cities in Malaysia

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**Abstract** The diurnal variations of ground-level ozone ( $O_3$ ) concentrations along a gradient of three major port cities (Klang, Perai, and Pasir Gudang) in Malaysia were evaluated. Annual monitoring records (2009) of  $O_3$ , nitrogen dioxide ( $NO_2$ ), nitric oxide (NO), carbon monoxide (CO), sulfur dioxide ( $SO_2$ ), and particulate matter ( $PM_{10}$ ) were obtained. Weather-influencing parameters such as incoming solar radiation, temperature, relative humidity, and wind speed were also analyzed. Although an overwhelming majority (99.9 %) of the recorded data are below the prescribed levels in the Malaysian Ambient Air Quality Guidelines (MAAQG), a number of exceedances (0.1 %) ( $>100$  ppb) have been recorded in Klang and Pasir Gudang. The annual hourly average concentrations of  $O_3$  were  $20.3 \pm 18.2$ ,  $15.4 \pm 15.8$ , and  $14.4 \pm 13.1$  ppb for Klang, Perai, and Pasir Gudang, respectively. The diurnal cycles of ground-level  $O_3$  concentrations and cluster analysis suggest that a unimodal peak occurs between 1 p.m. and 3 p.m., and the highest  $O_3$  concentrations were observed during the first quarter of 2009 as a result of the higher rate of local photochemical production. Results of principal component analysis showed that the contributions to  $O_3$  level variation by meteorological variables (UVB, temperature, relative humidity, and wind speed) are higher at 47.7, 39.5, and 18.2 % than those of primary air pollutants ( $NO_x$ , CO,  $SO_2$ ,  $PM_{10}$ ) at 28.9, 32.6, and 45 % for Perai, Pasir Gudang, and Klang, respectively. The exposure of the population in the three port cities to the elevated levels of ozone during January–May months validated the increasing threat and risks that ozone presents to humans. Result indicated that the concentration of  $O_3$  in the three ports was still below the maximum permissible values prescribed by the MAAQG, and expansion in

shipping industries is possible in exaggerating  $O_3$  problems especially in Klang.

**Keywords** Ozone · Diurnal variations · Malaysia

## Introduction

The trend of studies shifted increasingly in the last few decades from focusing on carbon- and sulfur-based air pollutants, such as carbon monoxide, carbon dioxide, and sulfur dioxide to become aware of tropospheric photochemical reactions and its capability to transform primary air pollutants into secondary air pollutants. In view of this scenario, ground-level ozone ( $O_3$ ), which is one of the major photochemical oxidants produced by these photochemical reactions, has gained prominence (Alghamdi et al. 2014).

At ground level,  $O_3$  is a major component of photochemical smog, and it has always been associated with negative impacts toward human health, vegetation growth, and materials lifetime.  $O_3$  is also considered a greenhouse gas that contributes to global warming (Reid et al. 2008; Alghamdi et al. 2014). Various studies have reported that  $O_3$  plays a significant role in tropospheric chemistry, because  $O_3$  is one of the principle precursors of hydroxyl radical (OH), which controls the oxidizing power in the lower atmosphere (Duenas et al. 2004; Singla et al. 2012). Ground-level  $O_3$  is produced by a series of complex photochemical reactions between its precursors, such as nitrogen oxides ( $NO_x$ ) or volatile organic compounds (VOCs), and incoming solar radiations (Seinfeld and Pandis 2006; Ghazali et al. 2010; Tsakiri and Zurbenko 2011).

Several studies reported that  $O_3$  exhibits strong diurnal variations which are controlled by various processes, including photochemistry, physical/chemical removal, and the rate of deposition and transport (Ghazali et al. 2010; Alghamdi

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# Emission of ozone precursors and particulate matter at signalized intersection of developing town

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**Key Words:** Vehicle emissions, level of service, delay time, ozone precursors

## ABSTRACT

This study aims to analyze and develop strategies on how to reduce the vehicle emissions at six signalized intersections during non-peak hours during construction period at Parit Buntar, Perak, by using Sidra Intersection 5.1. Vehicle emissions include pollutants such as CO, nitrogen oxide (NO<sub>x</sub>), hydrocarbon, and particulate matters. Both objectives are accomplished by analyzing the effect of signal phasing, cycle time, level of service, lane addition, provision of Left Turn On Red, and new approach at signalized intersections. The effects of vehicle emission on the environment were overlooked by traffic engineers because of their focus on the development of traffic signalized intersections. The more pressing issue is the rate of NO<sub>x</sub> converted to ground level ozone in the presence of sunlight (NO<sub>x</sub> = NO + NO<sub>2</sub>), with the main emitters of NO<sub>2</sub> being traffic and industries. Junction 4 is a congested and polluted signalized intersection with a released NO<sub>x</sub> of 80.7 kg h<sup>-1</sup>, whereas Junction 3 recorded 14.5 kg h<sup>-1</sup> NO<sub>x</sub>. The congestion factor, which is due to the rapid development of new construction work (flyover and double track), is influenced by the increasing total volume of vehicles from the three nearest towns within a 3-km radius between Parit Buntar, Nibong Tebal, and Bandar Baharu. A comparison of traffic congestion between 2011 (non-peak hours) and 2010 (peak hours) showed 100% increase in vehicle volume in five out of six signalized intersections.

## INTRODUCTION

Pollution from motor vehicles has become an important issue because of the increasing number of vehicles in use, distance travelled by vehicles, and signal delay time at intersections. Similar to population, the number of motor vehicles in Malaysia has been increasing rapidly since 2011, with a total of 21.4 million motor vehicles of all types in 2011. According to the Ministry of Transport Malaysia [1], motorcycles comprise the highest number of motor vehicles at close to 10 million, followed by motorcars at around 9.7 million.

At an optimum speed in excess of 60 km h<sup>-1</sup>, the emissions of CO<sub>2</sub> and oxides of sulfur (SO<sub>x</sub>) vary directly with fuel consumption. Chen et al. [2] concluded the tailpipe emissions of CO, nitrogen oxide (NO<sub>x</sub>), particulate matter, and hydrocarbons

(HC) vary with engine design, air-to-fuel ratio, and vehicle operating characteristics. With increasing engine temperature (or increasing vehicle speed), NO<sub>x</sub> emissions increase, whereas CO, particulate, and HC emissions decrease [3].

This paper focus on understanding vehicle emission at signalized intersections according to the effect of cycle time, level of service (LOS), and signal phasing on vehicle emissions. Hence, after analyzing the selected intersections in terms of changing signal phasing, cycle time, and LOS, mitigation was selected to overcome problematic traffic emissions. Reduction in traffic gaseous emission of NO<sub>x</sub> will reduce the production of ozone in ambient air [4].

A four-mode elemental model for estimating fuel consumption, operating cost, and pollutant emissions was generated by Sidra Intersection 5.1. For each lane of traffic, the program constructs drive cycles consisting

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## High Nighttime Ground-Level Ozone Concentrations in Kemaman: NO and NO<sub>2</sub> Concentrations Attributions

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### ABSTRACT

High nighttime ozone (O<sub>3</sub>) concentration levels were observed in Kemaman, Terengganu, and results were compared with those in other places in Malaysia. In this study, the contribution of precursors [nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>)] and meteorological factors wind speed, and wind direction) toward long-term high nighttime O<sub>3</sub> over the period of 1999 to 2010 was evaluated. During this period, the recorded highest nighttime O<sub>3</sub> ground level was 89 ppb with more than 25% surpassing 20 ppb. Analysis shows that minimal decreasing trends were measured in Kemaman. Lower nitrogen oxide (NO<sub>x</sub>) concentrations restricted the sinking agents; thus, reducing the depletion rates allowed O<sub>3</sub> to remain in the atmosphere. Minimal associations were observed between the O<sub>3</sub> concentration level and the speed and direction of wind. Accordingly, the largest contributor toward high nighttime O<sub>3</sub> ground level concentration in Kemaman was most probably NO<sub>x</sub> concentration.

**Keywords:** Long-term variations; NO<sub>x</sub> concentration; Nighttime ozone chemistry.

### INTRODUCTION

Ozone (O<sub>3</sub>) is an important constituent of air that plays significant roles as an oxidant and greenhouse gas. Most of O<sub>3</sub> concentrations occupy in ozone layer which, is located in the middle of the stratosphere and acts as a shield that protecting Earth from harmful radiation. However, a small concentration of O<sub>3</sub> also exists in the troposphere. At the ground level, O<sub>3</sub> is considered as a secondary air pollutant, which is typically associated with degrading air quality, and induces harmful effects on human health, crop production, material quality, and the ecosystem. O<sub>3</sub> is emitted from various anthropogenic activities. In particular, the formation and accumulation of O<sub>3</sub> are induced by the emissions of nitrogen oxide (NO<sub>x</sub>) and volatile organic compounds (VOCs). At the ground level, NO<sub>x</sub> plays an important role in the O<sub>3</sub> chemistry (Seinfeld and Pandis, 2006). In the presence of sunlight, nitrogen dioxide (NO<sub>2</sub>) undergoes photochemical reactions to produce free oxygen atom (O), which mainly reacts with oxygen molecule (O<sub>2</sub>) to form O<sub>3</sub> (Duenas *et al.*, 2004; Abdul-wahab *et al.*, 2005; Ghazali *et al.*, 2010). The variation in O<sub>3</sub> concentrations is influenced by meteorological factors. Such variation acts as a catalyst

of photochemical reactions as well as a mixing and dispersion agent.

O<sub>3</sub> has received substantial attention worldwide because of its negative effects. Ghosh *et al.* (2013) reported that background O<sub>3</sub> has increased over the last decade and is expected to continuously increase in the subsequent years. Many countries, including Malaysia, monitor the current O<sub>3</sub> condition and have set guidelines against this air pollutant. Considerable research employed the variability of O<sub>3</sub> concentrations on daily, seasonal, weekend, and weekday bases. The variability of O<sub>3</sub> concentrations in urban (Alvarez *et al.*, 2000; Ghazali *et al.*, 2010; Hassan *et al.*, 2013), suburban (Reddy *et al.*, 2011; Swamy *et al.*, 2012), and rural (Duenas *et al.*, 2004) conditions were also described by several studies. Toh *et al.* (2013) and Ghozdic *et al.* (2011) explored the role of meteorology in the variation of O<sub>3</sub> concentrations and concluded that O<sub>3</sub> is significantly responsive toward temperature, UV radiation, relative humidity, as well as wind speed and direction. Intense solar radiation, high temperature, minimal rainfall, low wind speed, and low relative humidity can raise O<sub>3</sub> concentration (Toh *et al.*, 2013). Substantial research also realized the possibilities of adopting multivariate analysis to produce prediction and forecasting models of O<sub>3</sub> concentrations (Pires *et al.*, 2008; Kovac-Andric *et al.*, 2009; Jimenez-Hornero *et al.*, 2010; Ghozdic *et al.*, 2011). Ishi *et al.* (2007) and Mohammed *et al.* (2013) examined the phytotoxic effects of O<sub>3</sub> on crop production. At present, the number of studies that have investigated the variations

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## Multivariate methods to predict ground level ozone during daytime, nighttime, and critical conversion time in urban areas

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### ABSTRACT

Ground-level ozone ( $O_3$ ) is known to exhibit strong daily variations that lead to complexity of the pollutants' analysis and predictions. This study aimed to introduce and explore the variations in  $O_3$  concentrations during daytime (DT), nighttime (NT), and critical conversion time (CCT) using multiple linear regression (MLR) and principal component regression (PCR) analyses. The original variables and principal component analysis (PCA) results were used as the input for MLR analysis. Hourly averages of six air pollutants and four meteorological parameters at Shah Alam during 1999–2009 were selected for this study. The monitoring records in 2010 were used to assess the developed models using several performance indicators. Results showed that the MLR model during DT exhibited optimal performance in terms of normalized absolute error, index of agreement, prediction accuracy, and coefficient of determination ( $R^2$ ) with values of 0.2762, 0.9211, 0.8581, and 0.7354, respectively. PCR during CCT also showed significantly higher performance than that during DT and NT. This result was evidenced by higher percentage of total variances, which could be explained by the selected variables in PCA during CCT.

**Keywords:** MLR, PCR, performance indicators,  $NO_2$  photolysis, NO titration



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### 1. Introduction

Ground-level ozone ( $O_3$ ), a secondary air pollutant from anthropogenic activities, is one of the critical air pollutants that is always associated with degrading air quality worldwide. It is also a greenhouse gas in both the stratosphere and troposphere (Chattopadhyay and Chattopadhyay, 2012). Exposure to  $O_3$  may harm human health via respiratory diseases, and lead to a decrease in lung function (Banan et al., 2013). In the presence of sunlight, nitrogen dioxide ( $NO_2$ ) undergoes photochemical reactions to produce free oxygen atom (O), which later reacts with oxygen molecules ( $O_2$ ) to form  $O_3$  (Duenas et al., 2004; Azmi et al., 2010). Once  $O_3$  is created, it is destroyed through several pathways, such as nitric titration and surface deposition (Abdul-Wahab et al., 2005).

The dependency of  $O_3$  formation toward UV light is associated with its clear daily variations. As a result, significant differences exist between daytime (DT) and nighttime (NT)  $O_3$  based on the diurnal plots (Han et al., 2011; Reddy et al., 2011; Ghosh et al., 2013). Various studies reported that the daily maximum  $O_3$  concentration is reached at late afternoon, similarly this value is reached at approximately 2 pm in Malaysia (Duenas et al., 2004; Azmi et al., 2010; Ghazali et al., 2010).  $O_3$  concentration variations show an interesting pattern in the morning because of the increment in  $O_3$  precursors emitted by various anthropogenic activities. Jimenez-Hornero et al. (2010) mentioned that  $O_3$  reaches the lowest concentration in the morning because of the higher rate of NO titration. Once the minimal point is reached,  $O_3$  starts to increase with rising  $NO_2$  concentration, thereby promoting  $NO_2$  photolysis. The point when the  $NO_2$  photolysis rate

is higher than the NO titration rate is very crucial in ground-level  $O_3$  chemistry because the difference in the chemical reaction's rate is expected to result in  $O_3$  accumulation. Toh et al. (2013) reported that air pollutants, such as  $O_3$ , are very responsive to changes in meteorological parameters. Thus, elevated  $O_3$  levels are often associated with intense solar radiation, high temperature, minimal rainfall, low wind speed, and low relative humidity.

The study of ozone variations is complex because of various possible precursors, photochemical processes, and meteorological factors (Chattopadhyay and Chattopadhyay, 2012; Toh et al., 2013). In addition, the interactions among  $O_3$ , its precursors, and meteorological parameters occur within a wide range of temporal and spatial scales (Abdul-Wahab et al., 2005). In explaining the variations in  $O_3$  concentration, various approaches have been applied. There are five multivariate techniques, such as multiple linear regression (MLR), principal component analysis (PCA), cluster analysis, Fourier analysis, and artificial neural networks which have been exploited to explain the variability in large air pollution data (Sousa et al., 2007; Ozbay et al., 2011; Dominick et al., 2012). In Malaysia, Ghazali et al. (2010) reported that the use of MLR in predicting  $O_3$  at an urban station is efficient and it is a highly useful tool in providing early information to the public. PCA, as a multivariate technique, has been used in ground-level ozone studies worldwide (Abdul-Wahab et al., 2005; Ozbay et al., 2011).

According to the literature, data on  $O_3$  prediction using PCA in tropical areas, such as Malaysia, are unavailable. The present study aimed to present the results of MLR analyses using the original variables and principal components (PCs) as the inputs for three different time periods, namely, DT, NT, and critical conversion time

**LAPORAN TEKNIKAL**

**THE FLUCTUATIONAL AND TRANSFORMATION  
CHARACTERISTICS OF PEAK GROUND LEVEL OZONE IN  
MALAYSIAN AMBIENT AIR**

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# RESEARCH OUTCOME

## THE FLUCTUATIONAL AND TRANSFORMATION CHARACTERISTICS OF PEAK GROUND LEVEL OZONE IN MALAYSIAN AMBIENT AIR

### Background of research

Air pollution is a major problem worldwide which is unlimited to urban and industrial areas only because air pollution at all scales, from local, regional, continental to global (Vallero, 2008). Once the pollutants were emitted from their sources, it can travelled miles from the origin driven by wind and under suitable conditions, some of pollutants such as nitrogen oxides (NO<sub>x</sub>) and hydrocarbons also react to create secondary pollutants.

The detrimental effect of air pollution towards human health, crop productions, material quality, and surrounding environment being the major reasons for efforts to understand and control measures. The effects of air pollutant to humans vary based on susceptibility factors such as age, nutritional status and predisposing conditions (Kampa and Castanas, 2008). Thus, asthmatic patients, children, allergic individuals and elderly people are the group of individuals that were at higher risk (Bernstein et al., 2004).

Urbanization process characterized by the increase in population, higher traffic density, changes in lifestyle and increase in energy demand has contributed to air pollution problem in Malaysia. Stationary sources such as power plant, industrial waste incinerators, dust emission from construction industry and quarries and open burning are also the major contributors towards the alarming state of air pollution in Malaysia (Dominick et al., 2012). Regulations to regulate air pollution in Malaysia is enacted under Environmental Quality (Clean Air) Regulations, 2014. The regulation regarded air pollutants as smoke, cinders, solid particles of any kind, gases, fumes, mist, odours and radioactive. Air pollution sources in Malaysia are mainly from anthropogenic emissions such as motor vehicles, industrial sites,

development activities, power generations, land clearing, open burning and forest fire (Mohammed et al., 2013).

Besides PM<sub>10</sub>, O<sub>3</sub> is considered as one of the criteria air pollutants in Malaysia due to favourable atmospheric conditions, high precursor emission from motor vehicles and industrial activities (DoE, 2013). Exposure to O<sub>3</sub> concentrations to a healthy human will result in a decreasing forced vital capacity (FVC) and forced expiratory volume (FEV), however, O<sub>3</sub> exposure towards asthmatic person will result in chronic inflammations in the lower airways (Bromberg and Koren, 1995; Berstein et al., 2004). Plants reacts to O<sub>3</sub> exposure by showing flecking, stippling, bleached, spotting and pigmentations (Ishii et al., 2007; Ainsworth et al., 2012; Mohammed et al., 2013). O<sub>3</sub> also attacks carbon double bonds (C=C) in rubber materials and under additional stress, the bonds will eventually break (Lee et al., 1996; Vallero, 2008).

Fluctuations in O<sub>3</sub> concentrations in any given location resulted from the combinations of formations, destructions, transport and deposition process (Vingarzan, 2004). The combined effect of precursors and meteorological conditions contribute to complex O<sub>3</sub> studies and analysis. In Malaysia, O<sub>3</sub> is scarcely studied by several researchers focusing in different aspects such as concentration variations (Latif et al., 2012; Banan et al., 2013); guidelines exceedances (Ahamad et al., 2014); phytotoxicity risk (Ishii et al., 2007; Mohammed et al., 2013); effect of meteorological parameters (Toh et al., 2013); and O<sub>3</sub> transformations and prediction (Ghazali et al., 2010). These studies address the O<sub>3</sub> problems on highly urbanized areas in the country which is the Klang Valley.

However, Klang Valley is characterized by high population and traffic density, major roads and highways, residential and commercial establishments and surrounded by industrial parks. This scenario does not represent the whole picture as there are places which showed low rates of development and less anthropogenic emissions. In addition, the period of these studies are in between one to six years. The earliest data from 1998 to 2001 have been presented by Ishii et al. (2007), while Ahamad et al. (2014) used data from 2008 to 2010. Meanwhile, those studies will give a clear picture in the trend of O<sub>3</sub> if longer duration is used i.e over a decade.

Photochemical reactions is undeniably the most significant path of O<sub>3</sub> formations at the ground level. Intensive work have been carried out by several studies in order to gain understanding on the mechanism behind the formations of O<sub>3</sub> (Atkinson, 2000; Jenkin and Clemitshaw, 2000; Atkinson and Arey, 2003). The dependency of the O<sub>3</sub> formation towards UV light is associated with its clear daily variations. In the presence of sunlight, nitrogen dioxide (NO<sub>2</sub>) undergoes photochemical reactions to produce free oxygen atom (O), which later reacts with oxygen molecules (O<sub>2</sub>) to form O<sub>3</sub> (Duenas et al., 2004; Azmi et al., 2010). O<sub>3</sub> concentration variations show an interesting pattern in the morning where O<sub>3</sub> level reaches the lowest concentration because of the higher rate of NO titration (Jiménez-Hornero et al., 2010). Once the minimal point is reached, O<sub>3</sub> starts to increase with the rise in NO<sub>2</sub> concentration, thereby promoting NO<sub>2</sub> photolysis. When the NO<sub>2</sub> photolysis rate is higher than the NO titration rate, critical conversion point (CCP) occurs. Therefore, CCP knowledge is very crucial in understanding ground-level O<sub>3</sub> chemistry because the difference in the chemical reaction's rate is expected to result in O<sub>3</sub> accumulation.

The literature review revealed that there are very limited studies on CCP where most of the studies were conducted only focusing on NO<sub>2</sub> photolysis rate (Clapp and Jenkin, 2001; Han et al., 2011). Moreover, various temporal, spatial and meteorological factors in Malaysia such as time of day, monsoonal changes, latitudes, clouds, and aerosols may influence the amount of UVB radiation received at the ground level. These will affect ground level O<sub>3</sub> formation and influence CCP formation. This type of study is limited and while other O<sub>3</sub> studies in Malaysia were rather focusing on variability of the pollutants (Azmi et al., 2010; Banan et al., 2013; Ahamad et al., 2014) than the other important characteristics of O<sub>3</sub>. Studies related to CCP are scarce and further analysis in this area is required.

The usage of statistical modelling is capable in providing additional information in O<sub>3</sub> formations and fluctuations behaviour. The relationship between O<sub>3</sub>, its precursors and meteorological conditions is frequently studied utilizing multiple linear regression (MLR) (Barrero et al., 2006; Ghazali et al., 2010). Some researchers also used a combination of MLR and principal component analysis (PCA) (Lengyel et al., 2004; Abdul-Wahab et al., 2005; Sousa et al., 2007; Özbay et al., 2011). In Malaysia, only Ghazali et al. (2010) have attempted to develop the O<sub>3</sub>

prediction models in urban areas using MLR. Even though good correlations were established between O<sub>3</sub> and NO<sub>2</sub>, the absence of NO in the analysis were unable to completely explain the O<sub>3</sub> formation reactions as NO also plays significant roles equivalent to the NO<sub>2</sub> concentrations. Meanwhile, none of O<sub>3</sub> prediction models using the combination of PCA and MLR (PCR) were reported in Malaysia. Studies revealed that the PCR model is excellent in avoiding multicollinearity problems in O<sub>3</sub> analysis (Lengyel et al., 2004; Abdul-Wahab et al., 2005; Kováč-Andrić et al., 2009) and could be an important tool in predicting the O<sub>3</sub> concentrations.

Encouraged by lack of detailed O<sub>3</sub> studies in Malaysia, the aims of this study are to critically investigate the fluctuational and transformational behaviour of ground level O<sub>3</sub> in Malaysia. This study is carried out using a long period of monitoring records (12 years) and considering all types of land use (industrial, urban, sub-urban and background) across Malaysia. This study has also extended and improvised the usage of O<sub>3</sub> production rates ( $J_{\text{NO}_2}/k_3$ ) of earlier studies (Clapp and Jenkin, 2001; Gerasopoulos et al., 2006; Han et al., 2011) by introducing the critical conversion point (CCP) of O<sub>3</sub> formations. CCP is then used as the baseline in determining the critical conversion time (CT). CT is a period that is important in O<sub>3</sub> photochemical productions. In addition, this research also provides next hour O<sub>3</sub> prediction models using MLR and PCR during different periods of time.

### **Scope of Study**

This study was designed to investigate and explore long term ground level O<sub>3</sub> fluctuational and conversion behaviour from its precursors in the Malaysian climate using graphical and multivariate techniques from January 1999 to December 2010 (12 years). 18 out of 52 continuous air quality monitoring stations operated by Alam Sekitar Malaysia Sdn Bhd (ASMA) was selected in this study. The main criteria in selecting the studied locations are:

- a) percentage of captured records was more than 80% for the concentration of O<sub>3</sub>, NO<sub>2</sub> and NO

- b) to represent categorical distribution of monitoring sites, which are urban (seven stations), sub urban (two stations) industrial (eight stations) and background (one station)

The remaining stations that are not selected are mostly due to low percentage of captured data and unavailability of UVB data. This research utilized nine variables that were divided into two groups. One group comprised of air pollutants, such as ground-level ozone ( $O_3$ ), nitrogen dioxide ( $NO_2$ ), nitric oxide (NO), sulphur dioxide ( $SO_2$ ) and carbon monoxide (CO). The other group composed of meteorological parameters, such as temperature, relative humidity, wind speed, and incoming solar radiation. These variables were selected based on their relationships with  $O_3$ .  $NO_2$ , NO and CO as the principle precursors to  $O_3$  production (Clapp and Jenkin, 2001; Seinfeld and Pandis, 2006; Ahamad et al., 2014). Whereas, UVB is the main ingredient in  $O_3$  photochemical reactions and without UVB, the reactions could not be completed (Tiwary and Colls, 2009). The relationship between  $O_3$  and temperature is obtained through UVB and high relative humidity indicates wet or rainy conditions which can promote  $O_3$  scouring. Wind is a dispersion agent to any air pollutants in the atmosphere.  $SO_2$  are primary air pollutants that are being emitted from similar sources to  $O_3$  precursors (De Nevers, 2010). Increasing in this pollutant may provide indication of  $O_3$  increment due to excessive  $O_3$  precursor concentrations.

Fluctuational behaviours of the  $O_3$  concentrations are assessed during daily, diurnal (Duenas et al., 2004; Azmi et al., 2010; Ghazali et al., 2010; Banan et al., 2013) monthly and spatial using graphical analysis. The influence of  $NO_2$ , NO, temperature and incoming solar radiation towards  $O_3$  formations and variations are explored using principal component analysis (PCA) and composite diurnal plot. This study is the first to introduce and explore the possibilities of explaining the variation in  $O_3$  concentration using critical conversion point (CCP) of ground-level  $O_3$  formation in the study area. CCP is defined as the point when  $NO_2$  photolysis rate begin to surpass NO titration rate during morning which corresponds to positive ozone production rates determined using composite diurnal plot and differences in  $O_3$  production ( $J_{NO_2}$ ) and destruction rate ( $k_3$ ).

According to the literature, research on O<sub>3</sub> prediction using PCA in tropical areas, such as Malaysia, are unavailable. The present study aimed to present the results of MLR analyses using the original variables and principal components (PCs) as the inputs for four different time periods, namely, 'daily' (AT), daytime (DT), nighttime (NT), and critical conversion time (CT). In this research, DT is defined as the complete hour falling between sunrise and sunset (Clapp and Jenkin, 2001). In Malaysia, Mohammed et al. (2013) used 7 a.m. to 7 p.m. (12 h) as DT, and 7 p.m. to 7 a.m. as NT, while AT is a whole day from 12 a.m. for 12 p.m. (24 h). In contrast to DT and NT, CT was determined based on CCP.

MLR is selected because of their capabilities to predict the contributions of selected variables to O<sub>3</sub> variations (Abdul-Wahab et al., 2005). Meanwhile, the use of PCs as input in MLR is intended to increase the prediction accuracy by removing multicollinearity problem. Next hour O<sub>3</sub> prediction models is developed based on different group of monitoring stations. Group of monitoring stations by DoE is analysed and new groups are developed using ranking of means and cluster analysis (CA). Index of agreement (IA), prediction accuracy (PA) and coefficient of determination ( $R^2$ ) are used to measure the accuracy, while normalized absolute error (NAE), mean absolute error (MAE) and root mean square error (RMSE) are used to measure error in predicted O<sub>3</sub> concentrations.



## **OBJECTIVES**

This research was carried out to accomplish four main objectives:

- a) To determine the fluctuational behaviour of ground level O<sub>3</sub> and to investigate variation of O<sub>3</sub> exceedances using time series plot
- b) To determine the critical conversion point (CCP) of O<sub>3</sub> transformation from its precursors and to introduce O<sub>3</sub> critical conversion time (CT)
- c) To develop multiple linear regression (MLR) and principal component regression (PCR) models to predict O<sub>3</sub> concentration during 'daily' (AT), daytime (DT), nighttime (NT) and critical conversion time (CT) in different background criteria groups.
- d) To identify the optimal next hour prediction models in describing O<sub>3</sub> fluctuation in Malaysia using several performance indicators.
- e) To propose best control framework to manage the exposure and impact of GLO to public

## **METHODOLOGY**

### **INTRODUCTION**

All procedures applied in this study are depicted in research flow in Figure 1. The flow described the main criteria and procedures performed to achieve each objective. Analysis of fluctuational behaviour of O<sub>3</sub> in selected stations is started by conducting the descriptive analysis and followed by daily, diurnal, monthly and spatial analysis. Next, the influence of NO<sub>2</sub>, NO, temperature and UVB towards O<sub>3</sub> is assessed using graphical and multivariate techniques, thus lead to identify the CCP in each station. The procedure in developing new group of monitoring stations is also presented and followed by development of MLR and PCR models during AT, DT, NT and CT. Lastly, the equations for each performance indicators used in this study is presented.

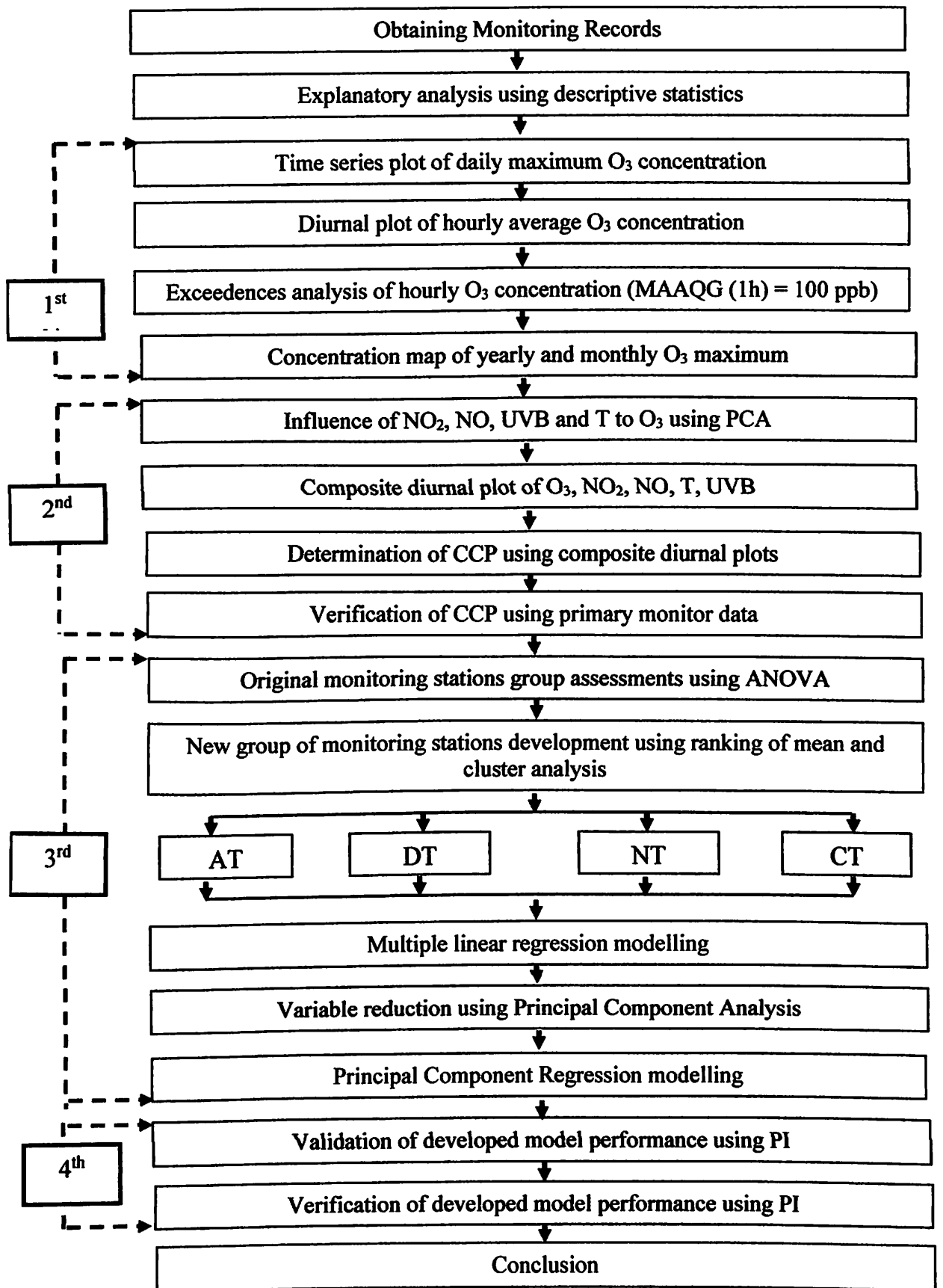


Figure 1 Flow of research methodologies

## **STUDY AREA AND MEASUREMENT TECHNIQUE**

The analysis in this study is carried out in 18 monitoring stations that represent urban, sub-urban, industrial and background areas across Malaysia. The hourly average O<sub>3</sub>, NO<sub>2</sub>, NO, SO<sub>2</sub>, CO, temperature, relative humidity, wind speed and incoming solar radiation from 1999 to 2010 were used.

### **Study areas**

The dispersion and dilution of air pollution are directly influenced by local attribution such as meteorological conditions as well as the location of the monitoring stations (Hosseinibalam et al., 2010). Location of the selected monitoring stations in Malaysia is illustrated in Figure 2 and description for each stations is shown in Table 1.

Bakar Arang monitoring stations is located in Kedah, a state in North Region of Peninsular Malaysia. It is located about 35 km to the north of Georgetown at intersection of two federal roads and federal highway improved the accessibility to the area, thus attracting high and medium business community. Main economy activities in Bakar Arang are manufacturing, semiconductors, and timber based industry (Noresah and Ruslan, 2009). In the vicinity of the monitoring station, there are a few industrial areas namely Tikam Batu Industrial area, Bakar Arang Industrial Area, Kedah Halal Park and Sungai Petani Industrial Area.

Perai and Seberang Jaya monitoring stations are located at Seberang Perai Penang, one of the most densely populated states in Peninsular Malaysia. Located in heavily industrial areas in Northern Region of Peninsular Malaysia, Seberang Perai covers an area about 738 km<sup>2</sup> that is divided into three districts which are Seberang Perai Utara, Tengah and Selatan. Both monitoring stations are located at Seberang Perai Tengah. There are approximately 20 industrial areas in Seberang Perai such as Perai Free Industrial Zone, Teguh Industrial Area, and Imperial Industrial Park etc. Meanwhile, the main economic activity that governed Seberang Jaya's economy are services and business.

Table 1 Description of selected monitoring stations

Group	Stations	Station ID	Specific Locations	Coordinate	Area (km <sup>2</sup> )	*Population
Industrial	Pasir Gudang	PG	SMK Pasir Gudang 2	N01°28.225' E103°53.637'	311.3	46,571
	Kemaman	KE	SK Bukit Kuang	N04°16.260' E103°25.826'	2,581	173,000
	Perai	PR	SK Taman Inderawasih	N05°22.265' E100°23.344'	738	362,820
	Kuching	KC	Medical Store	N01°33.734' E110°23.329'	1863	600,200
	Bukit Rambai	BR	Jalan Bukit Rambai	N02°15.924' E102°10.554'	468,8	474,500
	Ipoh	IP	SMK Jalan Tasek	N04°37.781' E101°06.964'	643	704,572
	Nilai	NL	SK Taman Semarak	N02°49.246' E101°48.877'	-	391,507
	Taiping	TP	SK Air Puteh	N04°53.940' E100°40.782'	186.46	217,647
	Urban	Kota Bharu	KB	Sultan Ismail College	N06°09.520' E102°15.059'	403
Kajang		KJ	Country Height	N02°59.645' E101°44.417'	92.98	342,657
Shah Alam		SA	SK TTDI Jaya	N03°4.636' E101°30.673'	290	541,306
Kota Kinabalu		KK	SMK Putatan	N05°53.623' E116°2.596'	351	423,300
Gombak		GB	Water Department	N03°15.924' E101°39.103'	628	681,300
Klang		KL	SMK (P) Raja Zarina	N03°00.620' E101°24.484'	636	832,600
Johor Bahru		KB	SMV Perdagangan	N01°29.815' E103°43.617'	1865	1,463,800
Sub Urban	Seberang Jaya	SJ	SK Seberang Jaya II	N02°49.246' E101°24.194'	738	362,380
	Bakar Arang	BA	SMK Tuanku Ismail	N05°37.886' E100°28.189'	930	175,609
BG	Jerantut	JT	Batu Embun Meteorological Office	N03°58.238' E102°20.863'	7,241	98,300

Note: \*2010; BG is background

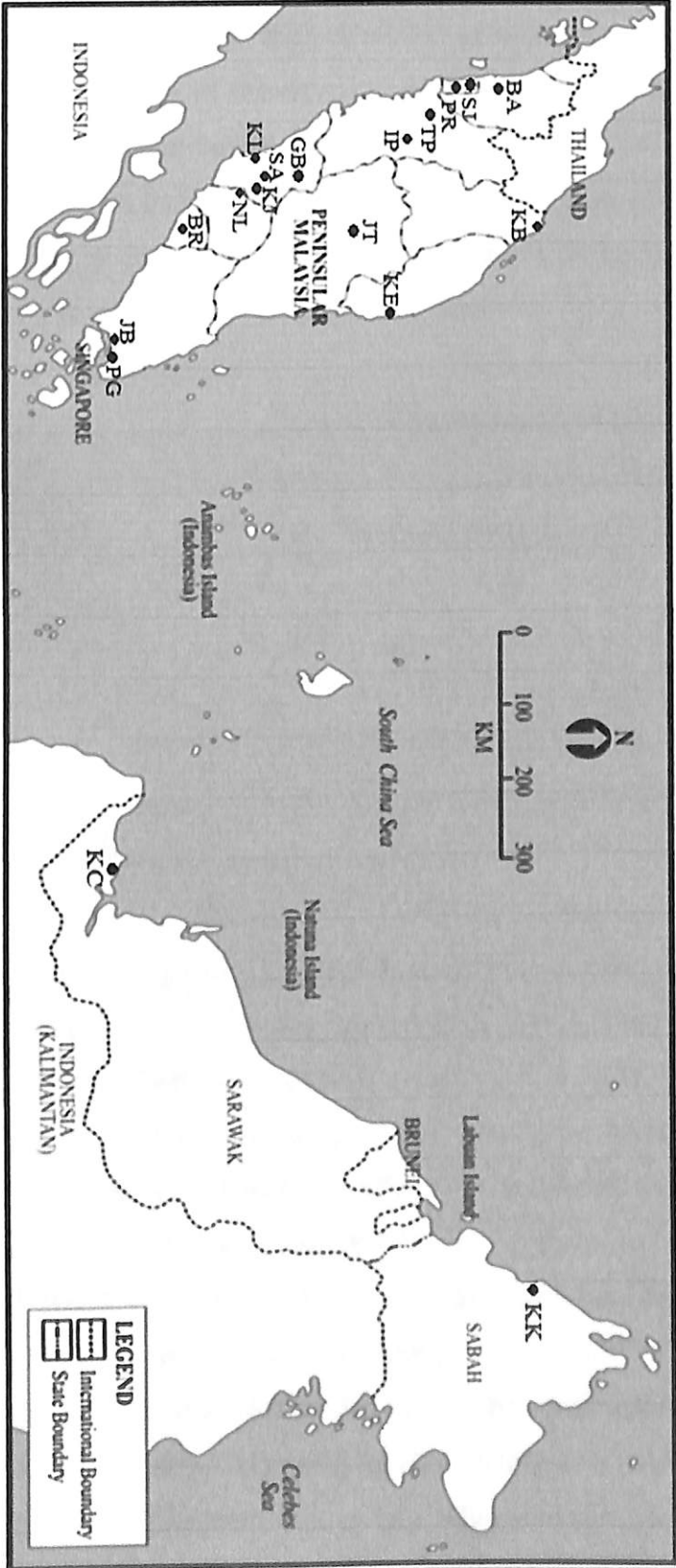


Figure 2 Specific location of monitoring stations across Malaysia (map is not up to scale)



Ipoh and Taiping are located at Perak, the second largest state in the Peninsular Malaysia. Ipoh is the capital city of Perak that is strategically located at Kinta Valley which is famous with tin mining area around Kinta River. The city is surrounded with limestones hills and Ipoh monitoring station is located approximately about 2 km radius from a cement factory. Ipoh is ranked as sixth populous city in Malaysia, even the population growth of the city is at negative rate (Malakahmad and Khalil, 2011). Taiping is the second largest city in Perak after Ipoh. Taiping was developed from mining and tin ore industries, however the development of the city is slowing down as its function as capital city of Perak was replaced by Kuala Kangsar and later was changed to Ipoh.

Shah Alam, Kajang, Gombak and Klang monitoring stations are located at Selangor, the most developed and heavily populated state in Malaysia. Shah Alam is a major city located about 25 km west of Kuala Lumpur, the capital of Malaysia. The city is served by six major highways, which experience heavy traffic in the morning and late afternoon rush hours (Azmi et al., 2010). Abdullah et al. (2011) reported that Shah Alam is a heavily industrialized area with high population and traffic density. The monitoring station is located in a large residential area near several industrial areas, such as Subang Industrial Park and Glenmarie Industrial Park (Azmi et al., 2010; Ghazali et al., 2010).

Kajang is located in the Langat River Basin, which is in the middle of Peninsular Malaysia (Latif et al., 2012). The city is located about 21 km from Kuala Lumpur and has a high level of commercial activity (Alahmr et al., 2012). The rapid growth in population, with a total population growth of 9 % per annum, has caused severe traffic congestion during the morning and afternoon rush hours (Alahmr et al., 2012). Klang is one of the districts in Selangor that is known as a port city. The rapid growth of Port Klang has greatly contributed to the developments of the Klang area. According to Haris and Aris (2013), Port Klang is the busiest port in Malaysia and the 14<sup>th</sup> 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 ports in Malaysia. Meanwhile, Gombak monitoring station is one of the earliest stations that have been established by Department of Environment, Malaysia to monitor urban air quality status. High-

populated residential areas and major roads surrounded the station, which experiences heavy traffic during morning and evening rush hour (Azmi et al., 2010).

Nilai monitoring station is located at Negeri Sembilan, a state that is located about 50 km to the south of Kuala Lumpur. Driven by industrial activities, Nilai is becoming one of the heavily industrialized areas in Southern Region of Peninsular Malaysia. The city has great accessibility through air and railways since the city is located near to Kuala Lumpur International Airport (KLIA) and Keretapi Tanah Melayu Railways Network. Nilai Utama Industrial Park is the main industrial area that is operated in Nilai (Ul-Saufie et al., 2013).

Bukit Rambai monitoring station is located at Malacca, the third smallest state in Malaysia after Perlis and Penang. The main economic activity in Bukit Rambai are timber processing and heavy industries.

Pasir Gudang monitoring station is located at Johor, the Southern State of Peninsular Malaysia. Pasir Gudang is one of the Johor's districts famous with Pasir Gudang Port. Port of Pasir Gudang is the fourth busiest port in Malaysia after the Ports of Klang, Penang, and Tanjung Pelepas (DoSM, 2012). In 2010, Port of Pasir Gudang handled about 28 million tons of cargo and received nearly 5000 international and local vessels (Ministry of Transport, 2010). These numbers increased in 2012, with the total number of departures and arrivals of vessels reaching up to 12,228. 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. Johor Bharu is the capital city for Johor. Other than highly populated residential areas and business premises, Johor Bahru also has a number of industrial areas.

Industrial station, Kemaman is located at east coast of Peninsular Malaysia at Terengganu. Kemaman is known for the discovery of petroleum near the town of Kerteh in the early-1980s. In general, Kemaman is relatively underdeveloped, except for a few places along the coastline where steel and petrochemical plants are located (Sulong et al., 2002). There are only two major industrial sites located in and near to

Kemaman, which are Kerteh Petrochemical Industrial Area and Gebeng Industrial Area (Ismail et al., 2011).

Kota Bharu monitoring station is located at Kelantan. Kota Bharu is Kelantan's capital city and located at the mouth of Kelantan River. 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).

Jerantut station is one and only background station that have been establish by Department of Environmental, Malaysia that is strategically located in the middle of Peninsular Malaysia. As a background station, this station is expected to be minimally affected by vehicular and industrial emissions. The station is surrounded by agricultural area and traditional Malaysian villages (Banan et al., 2013). According to Azmi et al. (2010), the source of the air pollution at Jerantut is expected to be natural forest, open burning, soil dust, and low number of motor vehicles.

Kuching monitoring station is located at northeast Bornea in Sarawak state. The station is situated in the centre of an industrial area that surrounded by congested roads during peak hours and close to South China Sea (Chung et al., 2012; Dominick et al., 2012) . In the vicinity of the monitoring station are Demak Laut Industrial Park, Kang Cheng Light Industrial Estate, Pending Industrial Estate and Sama Jaya Free Industrial Zone were situated.

Kota Kinabalu monitoring station is established in Sabah, East Malaysia. Kota Kinabalu is the capital city of Sabah that is located at the north of Borneo, which faces Celebes Sea to the east. The principle economy sector at Kota Kinabalu are industrial and tourism. Industrial areas such as Likas Industrial Area, Kolombong Industrial Area, Inanam Industrial Area and Kota Kinabalu Industrial Park are some of industrial that were established at Kota Kinabalu.

### **Climatic condition of study areas**

Climatically, Malaysia experience tropical rainforest climate distinguished by high temperature ranging from 22 to 24 °C during nighttime 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 to March) and the southwest monsoon (SWM) (June to September), and two intermoonson period occurred during April to May and October to November. Mean of the total annual rainfall in these studies areas 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.

### **Measurement techniques**

Continuous hourly air quality and meteorological parameters were obtained from the Air Quality Division of the Department of Environment, Ministry of Natural Resources and Environment of Malaysia. The obtained secondary data are regularly subjected to standard quality control processes and quality assurance procedures (Mohammed et al., 2013). The procedures used for continuous monitoring are in accordance with the standard procedures outlined by internationally recognized environmental agencies such as the United States Environmental Protection Agency (Latif et al., 2014).

#### **a) Ozone**

Teledyne UV Absorption Ozone Analyzer Model 400A is used to monitor hourly concentration of ground level O<sub>3</sub> (Ghazali et al., 2010). The analyzers have the EPA approval (EQOA-0992-087) (Teledyne, 2011) and subjected to schedule quality assurance and control (QA/AC) process of routine field calibrations and data variations test (Ishii et al., 2007). The analyser is ideal to be used in detecting O<sub>3</sub> concentration in the ranges of 0-100 ppb to 0-10 ppm at working temperature of 5°C to 40°C (Teledyne, 2011).

The analyzer applies a system based on the Beer–Lambert law to measure low ranges of O<sub>3</sub> concentration in ambient air and gaseous media (Ghazali et al., 2010; Latif et al., 2012; Mohammed et al., 2013). The ambient O<sub>3</sub> concentration is detected from the internal electronic resonance of O<sub>3</sub> molecules using absorption of 254 nm UV light emitted from an internal mercury lamp (Teledyne, 2011). Light from lamp shines down a hollow quartz tube that is alternatively filled with gas sample and scrubbed gas to remove O<sub>3</sub>. The calculation of O<sub>3</sub> concentration was based on the  $I/I_0$  ratio, which is the ratio of intensity of light passing through the scrubbed gas to sample gas. The Beer-Lambert equation to calculate the ozone concentration is show in equation (3.1).

$$C_{O_3} = -\frac{10^9}{\alpha l} \frac{T}{273^\circ K} \frac{29.92 \text{ inHg}}{P} \ln \frac{I}{I_0} \quad (3.1)$$

Where,  $I$  is intensity of light passed through the sample;  $I_0$  is intensity of light through sample free of ozone;  $\alpha$  is the absorption coefficient;  $l$  is path length;  $C_{O_3}$  is the concentration of O<sub>3</sub> (ppb);  $T$  is the sample temperature (°K);  $P$  is the pressure in inches of mercury.

Based on the equation, O<sub>3</sub> concentrations directly depends on intensity ratio and indirectly affected by temperature and pressure by changing the density of the sample. Effect of temperature and pressure were addressed by equation (3.1), which is included in the calculation and automatically done by the analyser (Teledyne, 2011).

### **b) Nitrogen Dioxide and Nitric Oxide**

Samples of hourly NO<sub>2</sub> and NO were collected using the NO/NO<sub>2</sub>/NO<sub>x</sub> analyzer model 200A (Ghazali et al., 2010). The analyzer applies the chemiluminescent detection principle to detect NO<sub>2</sub> concentration in ambient air. The advantages of chemiluminescence principle lies in their capability in detecting specific emissions of the desired pollutants and its insensitivity towards changes in surface properties (Fontijn et al., 1973). Teledyne (2008) claimed that the microprocessor technologies in the analyzer enhance the monitoring ability of pollutant concentrations. The

analyzer was used to provide sensibility, stability, and ease of use needed for ambient or dilution CEM monitoring (DoE, 2010).

The analyser was designed to directly measure concentration of NO and NO<sub>x</sub>, while NO<sub>2</sub> concentration is measured based on the calculations from the light intensity of the chemiluminescence gas reaction as follows:



Electronically excited (NO<sub>2</sub><sup>\*</sup>) molecules were produced once NO reacts with O<sub>3</sub> (3.2) and release their energy by emitting a photon to a lower energy level (3.3). From the equations, the light intensity that has been produced by the reactions was proportional to the NO concentration. The analyzer measures NO concentration by digitizing the signal from photomultiplier tube (PMT). When the direct readings were carried out, any NO<sub>2</sub> concentrations present in the inflow streaming gas were suppressed through a converter containing molybdenum (Mo) based on equation (3.4).



After measuring the NO concentrations, the analyzer will measure the total NO<sub>x</sub> concentration. NO<sub>2</sub> concentrations were calculated by subtracting NO concentration from NO<sub>x</sub> concentrations.

### c) Sulphur Dioxide

The instrument used to monitor the concentration of SO<sub>2</sub> is Teledyne API Model 100A/100E, with reference method number EQSA-0495-100. The ambient SO<sub>2</sub> concentrations were detected based on UV fluorescence detection methods, which allows detection, limit from 50 – 1000 ppb in the ambient temperature range of 5 to 40°C (Teledyne, 2006). SO<sub>2</sub> absorbs in the range of 190 – 230 nm UV energy that was relatively free of quenching by air and poly-nuclear aromatic (PNA) interferences. In the instrument, UV radiations produced by internal UV lamp passes



through a 214 nm bandpass filter and excites the SO<sub>2</sub> molecule and producing fluorescence. The produced fluorescence is measured by a PMT with a second UV bandpass filter. The fluorescent radiation impinging upon the PMT is directly proportional to concentration of SO<sub>2</sub>.

#### **d) Carbon Monoxide**

The instrument used to monitor the ambient concentration of CO is Teledyne API Model 300/300E. The analyzer measured concentration of CO using non-dispersive, infrared absorption method (Beer Lambert) with 0.5% precision and the lowest detection limit is 0.4 ppm (Latif et al., 2014). Concentration of CO is measured using empirical relationship that relates the absorption of light with the properties of material over a defined distance where the infrared radiation (IR) travel through a sample chamber containing the varying concentration of CO (Teledyne, 2009). The measurements of CO concentration in the sample chamber were then made by computing the ratio between the peaks measurements when IR was through high CO concentration chambers and reference gas chambers. The Gas Filter Correlation (GFC) wheel was installed in the instrument to avoid the interfering effect of various others gases and water vapour that were capable to absorb IR and effecting the CO concentration readings (Teledyne, 2009).

#### **e) Meteorological Parameters**

Meteorological parameters monitoring were carried out using Met One 062 sensor for temperature, Met One 083D sensors for relative humidity, Met One 010C sensor for measuring wind speed, Met One 020C sensor for wind direction and Scientech Model UV-S-290-T for UVB measurements. The Met One 062 temperature sensor is widely used to measure differential temperature ( $\Delta T$ ) in the range of -50°C to 50°C ( $\pm 0.05^\circ\text{C}$ ) applying multi element thermistor detection principle (Met One, 2010a). Meanwhile, the Met One 083D, relative humidity sensors is based on the capacitance change as one-micron thick dielectric polymer layers absorbs water molecules through a thin metal electrode and cause the capacitance change that were proportional to relative humidity (Met One, 2008).

The Met One 010C wind speed sensor is an accurate sensor that were capable to monitor horizontal wind speed using three lightweight cup anemometer that can be operated in range of 0 – 60 m/s ( $\pm 0.07$  m/s) (Met One, 2010b). In complimentary, the Met One 020C wind direction sensors provides azimuth data using lightweight aerofoil vane coupled with single precision potentiometer that mechanically measured the azimuth direction from  $0^\circ - 360^\circ$  ( $\pm 3^\circ$ ) (Met One, 2010b).

### Missing Monitoring Records

Missing data or incomplete data matrices that may arose from insufficient sampling procedures, errors in measurements or fault in data acquisition is not addressed in this study. No imputation methods were applied and any missing values that have encountered were directly omitted from the analysis. Table 2 showed percentage of missing values for hourly concentration of O<sub>3</sub>, NO<sub>2</sub> and NO at all stations. Since, no imputation method is utilized, monitoring stations were selected based on their percentage of captured data for O<sub>3</sub>, NO<sub>2</sub> and NO concentration. Only monitoring stations that have missing values less than 20% is considered in this study.

Table 2 Percentage of missing values for hourly concentration of O<sub>3</sub>, NO<sub>2</sub> and NO during 1999 to 2010

Station	Percentage of missing values		
	O <sub>3</sub>	NO <sub>2</sub>	NO
Pasir Gudang	4.2	6.7	8.2
Kemaman	2.3	6.9	5.8
Perai	12.2	8.5	8.6
Kuching	6.2	9.8	8.3
Bukit Rambai	2.9	3.5	3.6
Ipoh	2.3	2.6	2.5
Nilai	3.8	3.7	3.8
Taiping	2.7	4.4	5.8
Kota Bharu	4.0	4.4	4.4
Kajang	9.4	9.8	9.6
Shah Alam	3.4	4.3	4.3
Kota Kinabalu	6.1	14.6	8.0
Gombak	3.2	2.7	3.5
Klang	3.4	8.2	3.3
Johor Bahru	3.3	3.1	3.1
Seberang Jaya	7.0	5.8	4.9
Bakar Arang	5.8	3.1	5.1
Jerantut	5.4	6.7	6.6

## Descriptive Statistic Using Box and Whisker Plot

Descriptive analysis is the initial procedure to understand important information from a data set by collecting, organizing, summarizing and presenting the monitoring records. In this study the descriptive informations of the O<sub>3</sub> concentration at each stations is explored using box and whisker plot developed using SPSS version 22. The box and whisker plot is an explatory graphic, normally used to show the distribution of a dataset. Important criteria in decipher a box and whisker plot is illustrated in Figure 3 as follow where it is divided into two important parts; the box and whisker part.

The box parts summarize informations about shape, dispersion and centre of the data. The lower edge of the box represent the first quartile (Q1), while the upper edge is represented by the third quartile (Q3). Thus, the box represent the interquartile range (IQR = Q3-Q1) or 50% of monitoring records. Inside the box, there is line indicate median concentrations. The line extending above and below the box is called whiskers which indicate the lowest and highest value in the data set (excluding outliers).

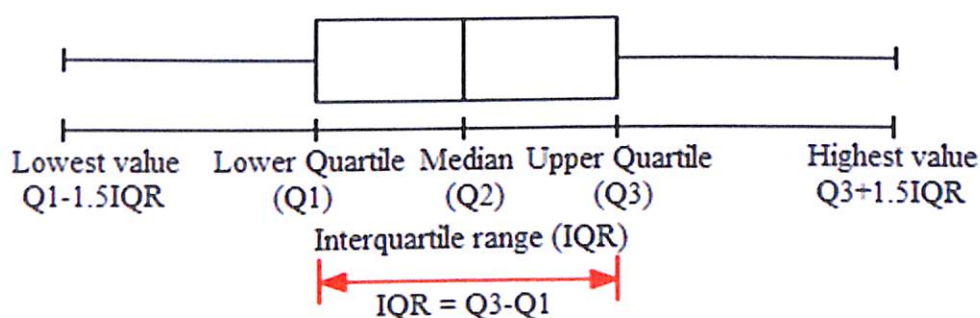


Figure 3 Definition of item in box and whisker plot (source: Govaerts et al., 1998)

## **ANALYSIS OF OZONE FLUCTUATIONAL BEHAVIOUR**

Fluctuational behaviours of ground level O<sub>3</sub> were analyzed during daily, diurnal, monthly and spatial in order to understand the characteristics and pattern of their fluctuations during studied period. In addition, the actual time of the occurrence high O<sub>3</sub> concentration above 100 ppb were also investigated.

### **Time Series Plot of Daily Maximum O<sub>3</sub> Concentrations**

The time series plots were used to show daily fluctuation behaviour of O<sub>3</sub> concentration and to detect the actual time where high O<sub>3</sub> concentration occurred. For the plots, daily maximum O<sub>3</sub> concentration were used. There are 365 data for each year except for leap year (2000, 2004, and 2008), which have 366 data. Through 12 year monitoring period, there are 4383 data of daily maximum O<sub>3</sub> concentrations for each respective locations.

### **Diurnal Plot of Hourly Average Concentrations.**

The diurnal plot is used to observe O<sub>3</sub> hourly fluctuation. Diurnal plots were constructed using 12 year hourly average data. Based on diurnal plot, hourly trends, time for maximum and minimum concentration and diurnal amplitude were determined. Diurnal amplitude is defined as differences in maximum and minimum concentration (Reddy et al., 2011). In this study, diurnal plots are also used to graphically explore variation in O<sub>3</sub> against NO<sub>2</sub>, NO, temperature and incoming solar radiation.

### **Exceedences Analysis**

The exceedences analysis were carried out to determine the number of hours that O<sub>3</sub> concentration exceed the MAAQG. One exceedences is defined as one data of 1-hour averaging O<sub>3</sub> concentration higher or equal to 100 ppb. 1-hour averaging limit of 100 ppb was used in the analysis instead of 8-hour averaging of 60 ppb in order to investigate hourly variations. The exceedences plots were then plotted for each stations to illustrate which stations that have more serious O<sub>3</sub> pollution problem,

while diurnal exceedences are also plotted to determine at which hour O<sub>3</sub> was exceeding the guidelines limit.

### **Ozone Concentration Map using GIS**

The usage of Geographical Information System (GIS) for inventory analysis, modelling and management of environmental events were relatively increasing (Narashid et al., 2010). Weng and Yang (2006) mentioned that GIS was proven to be the most appropriate and effective techniques in handling the spatial data such as air pollution, urban heat, land use cover and patterns. With the ability to perform analysis based on geographic location, GIS usage were able to assist in establishing and monitoring air quality and standards (Briggs et al., 1997). In this study, yearly and monthly maximum O<sub>3</sub> concentrations were used as locational attributes of the spatial data to generate concentration mapping using IDRISI: Selva Edition.

Surface interpolation techniques were used to generate O<sub>3</sub> concentration map for Malaysia. This interpolation techniques were sufficient to produce long-term pollutant concentrations map in an area that have minimal spatial variability (Briggs, 2005). Surface interpolation techniques were applied, where it calculates new values of O<sub>3</sub> concentration for unknown pixel using known pixels values of nearby point. Briggs (2005) mentioned that surface interpolation capability to determine values for nearby pixels provide ways for estimating pollutions levels at unmonitored sites and thus constructing the pollution maps. The 'Distance-Weighted Average', which is a one of the surface analysis, was selected in this study to interpolate the O<sub>3</sub> concentration value. According to Mohammed (2013), this technique capable to preserve the real O<sub>3</sub> concentration values at the monitored point and suitable to be adopted in O<sub>3</sub> studies. The interpolation function used in IDRISI is the inverse square distance ( $1/d^2$ , where d is distance).

In the first step, the coordinates of monitoring stations were converted into vector profile to determine the exact position for each station in map. Surface interpolation technique is then applied to the vector profile to produce a raster profile based on O<sub>3</sub> concentrations. After that, the interpolated O<sub>3</sub> concentration raster is 'overlay' with Malaysia base map to create the border to the map. Overlay process is the centre role



in IDRISI that needed two different raster maps to create a new map (Saeed et al., 2012). In the next step, the newly created O<sub>3</sub> map is then 're-class' using same interval from 1 ppb to 180 ppb to standardize all maps with different months and years. 180 ppb is choose as the maximum concentrations based on the highest O<sub>3</sub> concentration recorded during the studied period which is 176 ppb that was recorded in Kajang.

## **CRITICAL CONVERSION POINT OF OZONE FORMATION ANALYSIS**

The critical conversion points (CCP) of O<sub>3</sub> formation were investigated in this study using graphical and calculation of differences between rate of NO<sub>2</sub> photolysis and coefficient for the reaction of NO with O<sub>3</sub>. CCP is assumed as a starting point when NO<sub>2</sub> photolysis rate were larger than NO titration rate, thus begins the accumulation of O<sub>3</sub> concentration.

### **CCP Determination using Composite Diurnal Plot**

Graphically, CCP were determined based on composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, temperature and UVB radiation. CCP was assumed to occur at point of intersection between O<sub>3</sub>, NO<sub>2</sub>, and NO diurnal plots line. If the exact intersection point cannot be obtained from the plots, approximation of the interception point was point out as the CCP.

### **CCP Determination Based on O<sub>3</sub> Production Rate**

Seinfeld and Pandis (2006) claimed that the steady state of O<sub>3</sub> concentration is directly proportional to the [NO<sub>2</sub>]/[NO] ratio. Instead of using [NO<sub>2</sub>]/[NO], this study calculated the rate NO<sub>2</sub> photolysis over coefficient for the reaction of NO with O<sub>3</sub> ( $j_{\text{NO}_2}/k_3$ ) to indicate the variations in O<sub>3</sub> production rates during daytime (Clapp an Jenkin, 2001, Gerosopoulos et al., 2006; Han et al., 2011). The formula to calculate the value of  $j_{\text{NO}_2}/k_3$  is in equation (3.5).

$$\frac{j_{NO_2}}{k_3} = \frac{[O_3][NO]}{[NO_2]} \quad (3.5)$$

In this study, the differences in  $j_{NO_2}/k_3$  value at current hour ( $h_i$ ) to the previous hour ( $h_{i-1}$ ) denoted by  $\Delta j_{NO_2}/k_3$ . Positive differences of rates with previous hour indicating that  $NO_2$  photolysis rates is higher than NO titration rates, while negative indicate that  $NO_2$  photolysis is lower than NO titration. Positive value of  $\Delta(j_{NO_2}/k_3)$  indicate that  $O_3$  production is higher than destructions, while negative value show that  $O_3$  in decreasing trend. The biggest positive differences were used to point out the time for CCP.

### Verification of $O_3$ Critical Conversion Point

The existing of CCP is verified by re-visiting several selected locations. Air pollution and meteorological monitoring carried out at each selected location. The detail of the monitoring work in repective locations is depicted in the Table 3 as follows.

Table 3 Description of the verifications sampling stations

Stations	Sampling site	Monitoring period	Coordinate
Bakar Arang, Kedah	SMK Tunku Ismail	14-18 Oct 2014	N05°37.78' E100°28.067'
Nilai, Negeri Sembilan	SK Taman Semarak	4-8 Nov 2014	N02°49.275' E101°48.787'
Shah Alam, Selangor	SMK TTDI Jaya	8-12 Nov 2014	N 03°06.28' E101°33.374'

Monitoring of the  $O_3$  and  $NO_2$  concentrations were carried out using Aeroqual series 500. Aeroqual is a small handheld equipment with dimension of 195 x 122 x 54 mm with weight of 0.46 kg (Aeroqual Ltd, 2011). For monitoring  $O_3$  in the ambient air, gas sensitive semiconductors (GSS) sensor is used that allowed detection the presence of  $O_3$  concentration in the range of 0 to 150 ppb. Aeroqual Ltd (2011) reported that the sensor is capable to detect  $O_3$  concentration as low as 1 ppb with accuracy of calibration is  $\pm 5$  ppb and well operated in the range of temperature of 0 to 40°C and relative humidity of 10 to 90%. Meanwhile, monitoring for  $NO_2$

concentrations used gas sensitive electrochemical (GSE) sensor that can be used in the similar range of temperature and relative humidity with O<sub>3</sub> sensor. The detection range of the sensor is from 0 to 1000 ppb with calibration accuracy of ± 20 ppb.

The usage of Aeroqual for O<sub>3</sub> and NO<sub>2</sub> measurement enabling simultaneous data capture at lower net cost at relatively any desired locations. However, evaluation of the apparatus by Lin et al. (2015) found that Aeroqual series 500 only excel in O<sub>3</sub> monitoring while for nitrogen dioxide, unreliable result is produced. High correlation values ( $R^2 = 0.91$ ) is observed between O<sub>3</sub> that was monitoring using Aeroqual and UV-absorption O<sub>3</sub> analyser (Lin et al., 2015). In contrast, very weak correlation value ( $R^2 = 0.02$ ) is obtained from Aeroqual measurement and reference chemiluminescence NO<sub>2</sub> analyser. Bart et al. (2014) mentioned the NO<sub>2</sub> sensor is sensitive to water vapour and other pollutants concentration including ozone. In order to overcome the problem, Aeroqual technical support provide correlation factors as in equation (3.6) to calculate and correct NO<sub>2</sub> measurement.

$$[\text{NO}_2]_{\text{corrected}} = [\text{NO}_2] - [\text{O}_3] - 20 \text{ (*unit = ppb)} \quad (3.6)$$

Monitoring of ambient CO, CO<sub>2</sub> and TVOC were carried out using Indoor Air Quality Equipment (IAQ). The IAQ sensor probe dimension is 5 x 30 cm and operated using 2 x D cells. Each installed sensor were having different range where for CO<sub>2</sub> is from 0 to 10,000 ppm (±50 ppm), CO is from 0 to 500 ppm (± 50 ppm), TVOC is from 0.0005 to 20 ppm (± 0.0005 ppm). According to the Wolf Sense (2014) the sensor exhibited 90% response in a minute and would be able to operate in the temperature range of -15 to 60°C with relative humidity in the range of 0 to 90%. Meanwhile, the pocket PC used is SoMo 650 (windows mobile) which acted as the data logger to the IAQ set.

Meteorological parameters such as temperature, relative humidity, wind speed and direction were monitored using E-sampler. E-sampler is a particulate matter monitoring instrument enabled to measure near real time measurements. For temperature measurements, the instruments is enabled to measure temperature in the range of -30°C to 50°C. Meanwhile, the external relative humidity that were attached

to the instrument was able to measure the humidity of the air from 0 to 100%. For wind speed and direction, the instruments is capable in measuring wind ranged from 0.28 to 44.98 m/s, while the direction of 0 to 360°.

### **DEVELOPMENT OF NEXT HOUR OZONE PREDICTION MODEL**

Regression techniques are capable to investigate the relationship between a dependent variable and several independent variables. This techniques is used to develop O<sub>3</sub> prediction models using the original independent variables and significant principal components (PCs) from principal component analysis. Prediction models were developed during four different time period and because of these time periods are differ from one to other in term of time of the day and duration. Due to this, next hour prediction models were build in this study in order to take into account the spatial variability of O<sub>3</sub> concentrations during different periods of time.

#### **Daily, Daytime, Nighttime and Critical Conversion Time Determination**

O<sub>3</sub> was known to exhibit strong day-to-day variations (Ghazali et al., 2010). In the presence of sunlight and their precursors such as NO<sub>x</sub> and VOCs, O<sub>3</sub> was photochemically produced. In this study four time period were used as depicted in Table 4. The usage of different range of times is to explore the O<sub>3</sub> characteristics during different range of time. Lengyel et al. (2004) suggested that there are advantages of separating daytime and nighttime ozone concentration due to differences in influencing factors.

Table 4 Different time range used in prediction models

Time period	Duration (Hour)	Period
Daily (AT)	24	12 am – 12 pm
Daytime (DT)	12	7 am – 7 pm
Nighttime (NT)	12	7 pm – 7 am
Critical Conversion Time (CT)	4	8 am – 11 am

Daylight hours or daytime is defined as the complete hours falling between sunrise and sunset (Clapp and Jenkin, 2001). In Malaysia, daytime start from 7 am until 7 pm, and during 7 pm to 7 am are deemed as nighttime (Mohammed et al., 2013). Contrast to the daytime and nighttime, the critical conversion time was considered based on the photochemical reactions of O<sub>3</sub> formation. In this study, the critical conversion time (CT) is determined based on CCP considering the earliest and the latest of CCP. Only one CT will be consistently used for all stations.

### **Developing New Group of Monitoring Stations**

In this study, new group of monitoring stations is developed based on O<sub>3</sub> concentration utilizing two methods were used which are ranking of means and cluster analysis.

### **DoE Classification**

The original group based on the location of the monitoring stations that were established by Department of Environment, Malaysia was assessed using one-way analysis of variance test (ANOVA) using SPSS version 22. ANOVA is a widely used statistical test to estimate and test the hypothesis regarding the population means of a population. The null hypothesis of the test is the concentrations of O<sub>3</sub> from monitoring stations in the same group have the equal means. The significant level used is  $\alpha = 0.05$  (95% confidence level). *F* value is calculated using formula in Table 5 (Yahaya et al., 2008), meanwhile the critical values of F distribution is determine based on  $\alpha$ , degrees of freedom between monitoring stations and degrees of freedom in error.

Table 5 ANOVA formula

Source of variation	Sum of squares	Degrees of Freedom	Mean square	$F_o$
Between monitoring stations	$SS_R$	$k$	$MS_R = SS_R/k$	$MS_R/MS_E$
Error	$SS_E$	$n-p$	$MS_E = SS_E (n-p)$	
Total	$S_{YY}$	$n-1$		

Where  $SS_E$  is sum of squares due to error;  $SS_T$  is total sum of squares;  $SS_R$  is sum of squares due to regression;  $MS_E$  is mean square due to error;  $MS_R$  is mean square due to regression;  $k$  is number of monitoring stations;  $n$  is number of  $O_3$  monitoring records; The formula to calculate  $SS_E$ ,  $SS_T$  and  $SS_R$  is showed in equation (3.7), (3.8) and (3.9), respectively.

$$SS_E = \sum_{i=1}^n (y_i - \hat{y}_i)^2 \quad (3.7)$$

$$SS_T = \sum_{i=1}^n (y_i - \bar{y})^2 \quad (3.8)$$

$$SS_E = \sum_{i=1}^n (\hat{y}_i - \bar{y})^2 \quad (3.9)$$

Where  $y_i$  is  $O_3$  concentrations at station  $i$ ;  $\hat{y}_i$  is predicted  $O_3$  concentrations at station  $i$  and  $\bar{y}$  is mean  $O_3$  concentrations.

### Ranking of Means

Re-grouping the monitoring stations using the ranking of means was carried out using the calculations of means of the  $O_3$  concentration at one particular station using equation (3.10) during different time range (Yahaya et al., 2008).

$$\bar{x} = \frac{1}{n} \sum x_i = \frac{1}{n} (x_1 + x_2 + \dots + x_n) \quad (3.10)$$



Where  $\bar{x}$  is the mean of O<sub>3</sub> concentration;  $x_i$  is the hourly average O<sub>3</sub> concentration;  $n$  is the number of monitoring records. The calculated means were rank from lower to high concentrations and regrouped based on the range of the concentration as depicted in Table 6.

Uniform ranges with increment of 10 ppb were selected based on the mean concentrations of O<sub>3</sub> for each period of time. Range of 10 ppb was selected because study by Mustafić et al. (2012) used the same range to assess and quantify the association between short-term exposure to major air pollutants (ozone, carbon monoxide, nitrogen dioxide, sulfur dioxide, and particulate matter  $\leq 10 \mu\text{m}$  [PM<sub>10</sub>] and  $\leq 2.5 \mu\text{m}$  [PM<sub>2.5</sub>] in diameter) on myocardial infarction (MI) risk. Shah et al. (2015) also adopted same range to study the relationship between short term exposure to air pollution and they also claimed these increments were used in most studies that related to meta-analysis.

Table 6 Descriptions of newly developed group based on difference time range

Time range	Group	Range of O <sub>3</sub> Concentration (ppb)
Daily (AT)	G1-A	$10 \leq x < 20$
	G2-A	$20 \leq x < 30$
Daytime (DT)	G1-D	$10 \leq x < 20$
	G2-D	$20 \leq x < 30$
	G3-D	$x > 30$
Nighttime (NT)	G1-N	$0 \leq x < 10$
	G2-N	$10 \leq x < 20$
Critical Conversion	G1-C	$0 \leq x < 10$
Time (CT)	G2-C	$10 \leq x < 20$

### Cluster Analysis

Cluster analysis (CA) is a multivariate technique that primarily used in data reduction and summarization of voluminous observations records. CA is a method to categorize objects into a number of cluster/group, so that the objects with the cluster

are similar to each other while the objects in different cluster are different with each other's (Pires et al., 2008b; Lau et al., 2009). CA aims to create a group that maximize between group variance and to minimize within group variance (Lu et al., 2011).

The procedure for developments of new monitoring stations groups using cluster analysis is shown in Figure 4. The CA is performed using hourly average O<sub>3</sub> concentration during four time periods (AT, DT, NT, and CT). Hierarchical agglomerative cluster is selected to be used in this study to provide intuitive similarity relationship between O<sub>3</sub> concentration at one station to entire stations (Shrestha and Kazama, 2007).

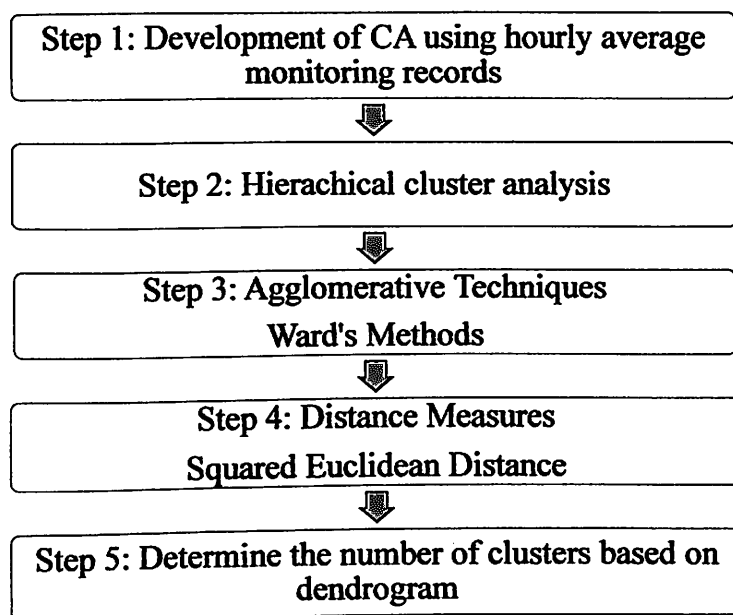


Figure 4 Procedure of developing new groups of monitoring stations using cluster analysis.

Meanwhile, Wards method is chosen to be used as agglomerative method because Wards method used an analysis of variance approach to evaluate the distance between clusters in attempt to minimize the sum of square (SS) of any two cluster (Shrestha and Kazama, 2007). Even though, the Euclidean Distance is the most familiar and commonly used distance measured, the squared Euclidean Distances measure is chosen in the study since there are large distance corresponded to many dissimilar items were expected to occur in O<sub>3</sub> analysis. The same agglomeration

technique and distance measure has been used by several researchers in air and water pollution studies (Shresta and Kazama, 2007; Lau et al., 2009). The formula of squared Euclidean distance between  $i$  and  $j$  is as showed in equation (3.11).

$$d_{ij}^2 = \left[ \sum_{k=1}^p (X_{ij} - X_{jk})^2 \right] \quad (3.11)$$

Where  $X_y$  is the value of the  $k^{th}$  variable for the  $i^{th}$  entity.

The classifications of the objects in cluster analysis were illustrated by dendrogram (tree diagram) which showed the similarity measured or the distances between any two variables. The numbers of the significant cluster were determined based on the biggest drop in the coefficients of Ward linkage.

### **Principal component analysis (PCA)**

In this study, PCA is used to test the contribution of selected variables to variation in  $O_3$  concentration as well as to reduce the number of variables to be used in MLR. The procedure of PCA analysis is shown in Figure 5. Hourly average monitoring records for 12 years period was used in the analysis. The analysis were carried out using IBM SPSS statistical software version 22.

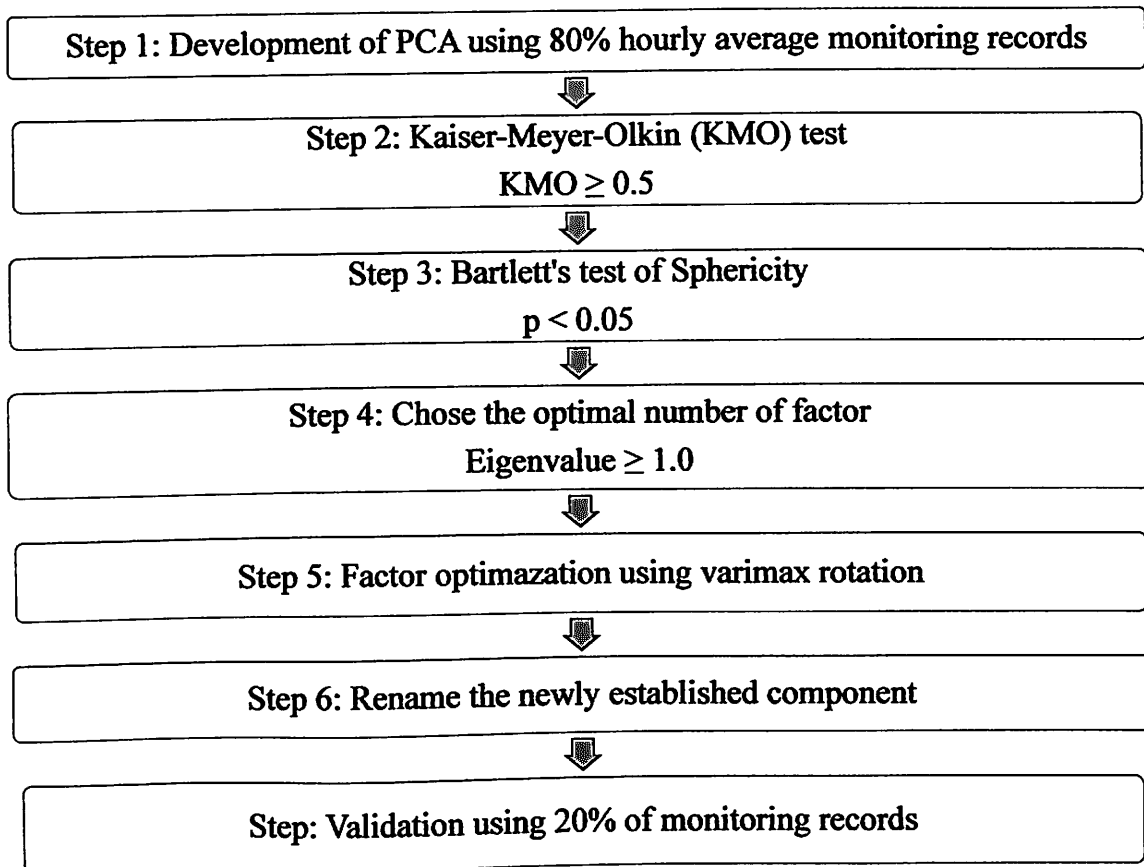


Figure 5 Development of principal component analysis procedure

The sufficiency of the monitoring records for PCA were assessed using Kaiser-Mayer-Olkin Measure (KMO) and Bartlett's Test of Sphericity. KMO measure is an index that compared the magnitudes of observed correlation coefficients with the magnitudes of the partial correlation confidents. Chattopadhyay and Chattopadhyay (2011) mentioned that KMO could give an indication whether PCA is suitable for removing multicollinearity in the monitoring records. KMO value close to 1 indicated that the correlation pattern is relatively compact and suitable for PCA, while when KMO value were close 0 gives the indication that the data were not suitable for PCA. The equation for KMO measure is given in (3.12).

$$KMO = \frac{\sum_{i \neq j} \sum_{i \neq j} r_{ij}^2}{\sum_{i \neq j} \sum_{i \neq j} r_{ij}^2 + \sum_{i \neq j} \sum_{i \neq j} a_{ij}^2} \quad (3.12)$$

Where  $r_{ij}$  is the simple correlation coefficients and  $a_{ij}$  is the partial correlation coefficients between  $i$  and  $j$ .

Typically, KMO is combined with Bartlett's test of sphericity ( $p < 0.001$ ) in determine the adequacy of data for PCA (Özbay et al., 2011; Ul-Saufie et al., 2013). According to Sousa et al. (2007), the null hypothesis in PCA is the variables are uncorrelated and to justify the hypothesis and applicability of the monitoring records for PCA, Bartlett's sphericity test were used. The equation for the test is expressed in equation (3.13).

$$x_k^2 = \left[ n - k - \frac{2(p-k) + 7 + \frac{2}{(p-k)}}{6} + \sum_{j=1}^k \left( \frac{\bar{\lambda}}{\lambda_j - \bar{\lambda}} \right)^2 \right] x \left[ -\ln \prod_{j=k+1}^p \lambda_j + (p-k) \ln \bar{\lambda} \right] \quad (3.13)$$

Where  $n$  is the numbers of observations;  $\lambda_j$  denotes the eigenvalue for  $i^{th}$  component; and  $p$  is the number of components. The following equation (3.14) can be used to calculate  $\bar{\lambda}$

$$\bar{\lambda} = \sum_{j=k+1}^p \frac{\lambda_j}{p-k} \quad (3.14)$$

In most of cases, only total variance higher than 60% is considered significant (Rajab et al., 2013). The PCs were extracted so that the first principal component (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 (Kovač-Andrić et al., 2009). Rajab et al. (2013) claimed that most of the variations in the data is can be described by the first few PC and relatively relevant in represent the original variables. PCs are generally expressed in (3.15).

$$PC_1 = l_{1i}X_i + l_{2i}X_2 + \dots + l_{mi}X_m \quad (3.15)$$

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

Özbay et al. (2011) stated that PCA uses eigenvalue of the covariance matrix which is a set of special scalars associated with linear system of the equation. An

eigenvalue greater and equal to one for principal component is commonly considered as being statistical significant (Lu et al., 2011; Ozbay et al., 2011) and in order to explain most of the variation in dataset (Elbayoumi et al., 2014). The equation for eigenvalue denoted in equation (3.16).

$$|c - \lambda I| = 0 \quad (3.16)$$

Where  $c$  is the correlation of the monitoring records;  $\lambda$  is the eigenvalue; and  $I$  is the identity matrix.

According to Kovač-Andrić et al. (2009), the PCs are extracted in ascending order, where the first component (PC1) represents the largest variation in the dataset. In common practice, application of PCA procedures is followed by orthogonal rotations such as varimax rotations (Lau et al., 2009; Dominick et al., 2012). PCs is subjected to rotation in order to gain better understanding and interpretation of the data. The advantage of rotation is also to optimize the components structure and equalized the relative importance of significant components. Varimax rotation ensure each variable is maximally correlated to only one PC and insignificantly related to other PC (Dominick et al., 2012). The factor loading after rotation is crucial in determine significant variable in each PCs. The factor loadings greater than 0.5 are considered strong, 0.4 are moderate, and 0.30 are weak. Ul-Saufie et al. (2013) suggested that only factor loading higher than 0.4 is considered significant in analysis, while weaker loading which is less than 0.4 should be suppressed from analysis. The last step in PCA analysis is to give an appropriate name to the principal components based on the variables with significant loadings.

### **Multiple Linear Regressions (MLR)**

Regression-based technique was used in this study because of their capabilities to predict the contributions of selected variables to O<sub>3</sub> variations (Abdul-Wahab et al., 2005). MLR used least squares method to fit the model such that the sum of squared differences between observed O<sub>3</sub> concentrations and predicted O<sub>3</sub> concentrations is minimized. The general equation of multiple linear regressions is expressed in (3.17) (Yahaya et al., 2008):

$$y_i = \beta_0 + \beta_1 x_{1i} + \beta_2 x_{2i} + \dots + \beta_k x_{ki} + \varepsilon_i \quad (3.17)$$

Where  $y_i$  is the dependent variable;  $x_{1i}, x_{2i}, \dots, x_{ki}$  are the independent variables;  $\beta_0, \beta_1, \dots, \beta_k$  are the coefficient of the regression model and  $\varepsilon_i$  is the random error term.

The steps in developments of MLR models are illustrated in Figure 6.

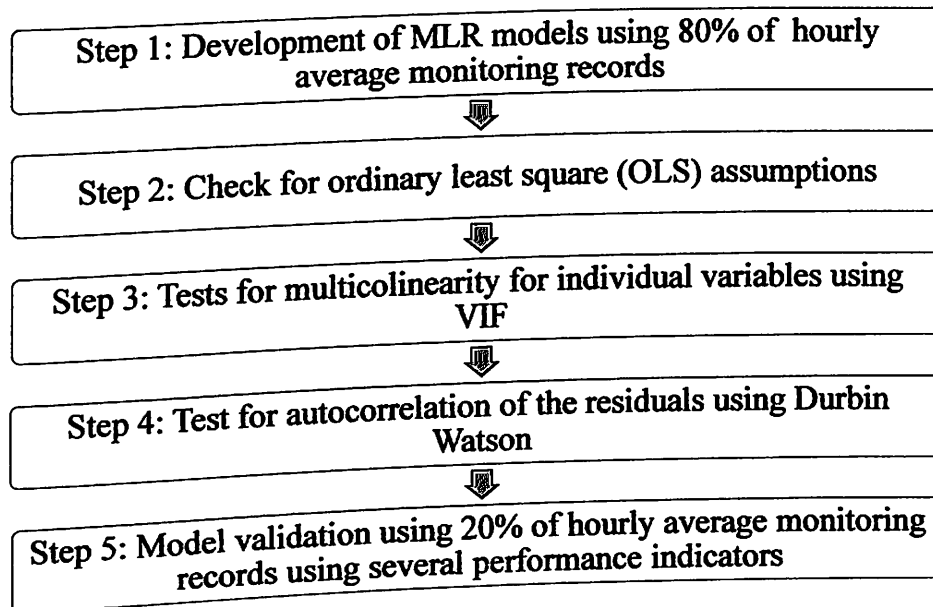


Figure 6 Procedure of developing MLR models during the predetermined time range

The MLR models during the ranges of time were developed using approximately 80% of the hourly average monitoring records, while the remaining 20% were used as the validations data. MLR models required several assumptions regarding the residual (error) such as zero mean, constant variance, normally distributed and uncorrelated with the independent variables (Ul-Saufie et al., 2013).

Variance Inflation Factor (VIF) is used to give an indication regarding the multicollinearity on the variance of the estimated regression coefficients. Arshad et al. (2013) mentioned that large VIFs value indicated multicollinearity problems and in practice, VIF of 10 is adopted as the critical threshold. Once the VIFs are exceeding the critical threshold value, gives the indication that the associated regression coefficients are poorly estimated due to multicollinearity problems. The formula for VIF is depicted in equation (3.18).



$$VIF_i = \frac{1}{1 - R_i^2} \quad (3.18)$$

Where  $VIF_i$  variance inflation is factor and  $R_i^2$  is the coefficients of determination in a regression of the  $i^{th}$  independent variables.

Meanwhile, the indication of the autocorrelation problems in residuals of MLR models were determined using Durbin Watson ( $d$ ) test and the formula is shown in equation (3.19).

$$d = \frac{\sum_{i=1}^n (\hat{e}_i - \hat{e}_{i-1})^2}{\sum_{i=1}^n \hat{e}_i^2} \quad (3.19)$$

Where  $n$  is the number of observations,  $\hat{e}_i = y_i - \hat{y}_i$ ;  $y_i$  are observed values;  $\hat{y}_i$  is predicted values.  $d$  values is explained based on the upper ( $d_U$ ) and lower ( $d_L$ ) critical values (Montgomery et al., 2012). If  $d > d_U$  indicate there no autocorrelation exist; if  $d < d_L$  indicate positive autocorrelations, if  $d_L < d < d_U$ , the test is inconclusive

Lastly, the developed models validated using 20% of the monitoring records using several performance indicators for errors and accuracy measures.

### **Principal Components Regressions (PCR)**

Principal Component Regressions (PCR) is a developed method by combining MLR and PCA. The usage of the PCs as the input in MLR is intended to reduce the complexity and multicollinearity problems of the models. The selected variables with high loading from PCA ensured that the majority of the original variances were included in the models and ideal to be used as independent variables in MLR (Gvozdic et al., 2011). The architecture of PCR model and the procedures in development of PCR models illustrated in Figure 7 and 8, respectively. The development of PCR models involved two stages, which are the PCA and MLR. During first stage, the selected independent variables will be analyzed using PCA and the output of PCA will be used as the input to MLR models that will gives the PCR models.

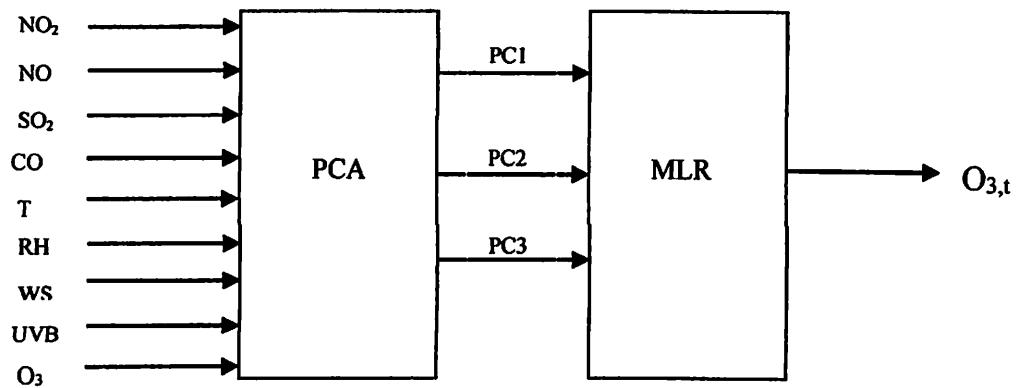


Figure 7 Architecture of a PCR models using PCA outputs as the input to MLR

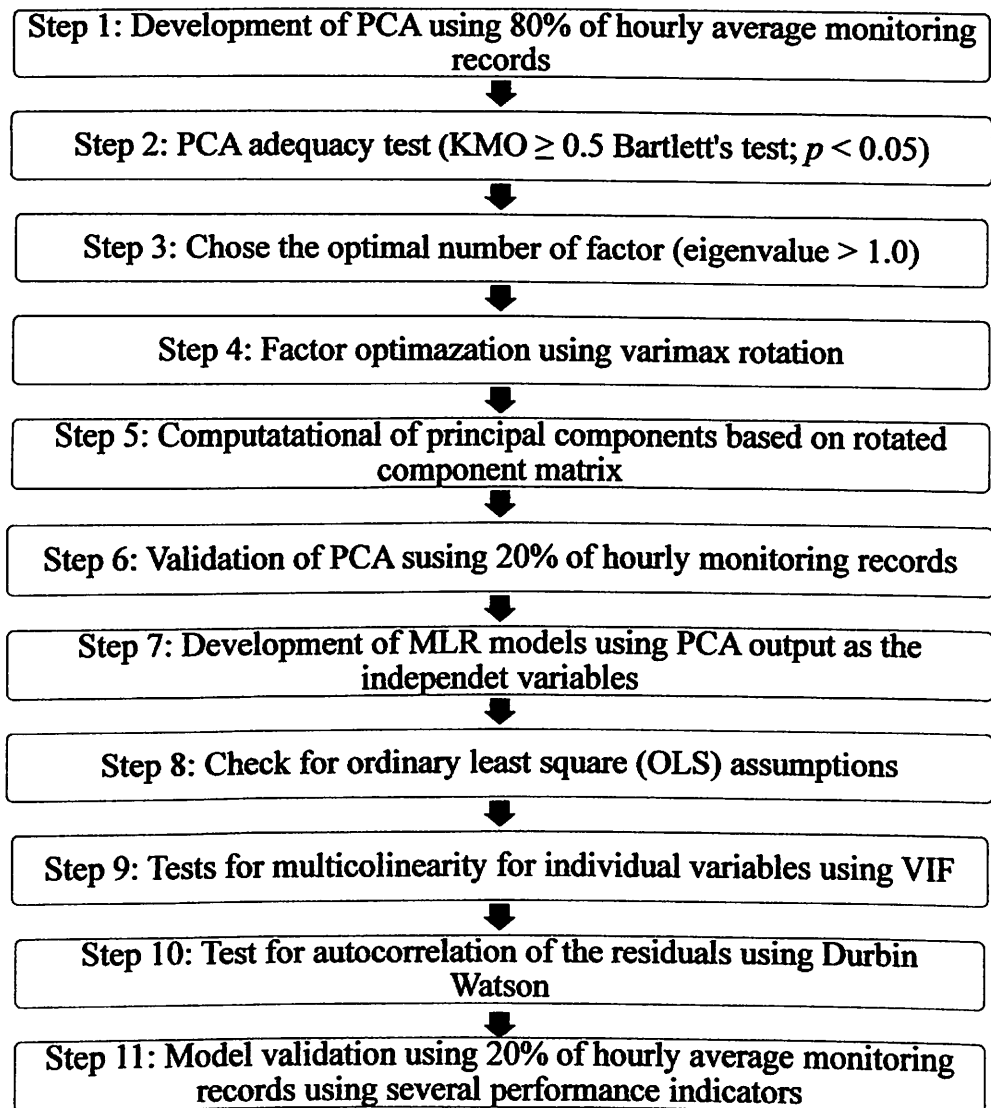


Figure 8 Procedure of developing PCR models during the predetermined time range

## MODEL VALIDATION AND VERIFICATION

The validation of the developed models were carried using approximately 20% of the total monitoring records. Meanwhile, the developed models were verified using newly measured data from several monitoring stations. Bakar Arang, Nilai and Shah Alam were selected to verify sub urban, industrial and urban models, respectively. The details of monitoring stations locations, duration, monitoring equipment's and techniques for O<sub>3</sub>, NO<sub>2</sub>, CO and meteorological parameters was described previous section (3.4.3). However, due to limitation in monitoring equipments for NO, SO<sub>2</sub>, and UVB, the value used in the verification stage is imputed from DoE dataset. These values is imputed by using the data at the same date as monitoring date at each station as mentioned in Table 3.3 during different year from 1999 to 2010.

The developed model's accuracy and errors measure was validated and verified using several performance indexes namely normalized absolute error (NAE), mean absolute error (MAE), index of agreement (IA), prediction accuracy (PA), root mean square error (RMSE) and coefficient of determination (R<sup>2</sup>). A good prediction model are indicated by high accuracy (closer to 1 for IA, PA and R<sup>2</sup>) and minimal errors (closer to 0 for NAE and RMSE) (Md Yusof et al., 2010; Mohammed et al., 2013; Ul-Saufie et al., 2013). The equations used to calculate the performance indexes are given in equations (3.20) to (3.24).

### Normalized Absolute Error (NAE)

NAE measures the average difference between predicted and observed values in all cases divided by observed values (Elbayoumi et al., 2014). NAE is expressed as follows:

$$\text{NAE} = \frac{\sum_{i=1}^n |P_i - O_i|}{\sum_{i=1}^n O_i} \quad (3.20)$$

where  $n$  is the total number of sample,  $P_i$  is the predicted concentration of ground-level O<sub>3</sub>, and  $O_i$  is the observed value of O<sub>3</sub> concentration.

### Mean Absolute Error (MAE)

MAE is used to measure the difference between observed and predicted values and the formula to calculate MAE is as follows:

$$\text{MAE} = \frac{1}{n} \sum_{i=1}^n |P_i - O_i| \quad (3.21)$$

where  $n$  is the total number of sample,  $P_i$  is the predicted concentration of ground-level  $O_3$ , and  $O_i$  is the observed value of  $O_3$  concentration

### Root Mean Square Error (RMSE)

RMSE is used to measure the success of numerical predictions. It can provide error values with the same dimensionality as actual and predicted values, and summarize between the observed and predicted values. Ul-Saufie et al. (2013) defined RMSE as follows:

$$\text{RMSE} = \sqrt{\frac{1}{n-1} \sum_{i=1}^n (P_i - O_i)^2} \quad (3.22)$$

where  $n$  is the number of sample,  $P_i$  is the predicted  $O_3$  concentration, and  $O_i$  is the observed  $O_3$  concentration.

### Index of Agreement (IA)

IA, a measure of accuracy, was calculated using equation (3.22) (Md Yusof et al., 2010).

$$\text{IA} = 1 - \left[ \frac{\sum_{i=1}^n (P_i - O_i)^2}{\sum_{i=1}^n (|P_i - \bar{O}| + |O_i - \bar{O}|)^2} \right] \quad (3.23)$$

where  $n$  is the total number of sample,  $P_i$  is the predicted  $O_3$  concentration,  $O_i$  is the observed  $O_3$  concentration, and  $\bar{O}$  is the mean of the observed  $O_3$  concentration.

### Prediction accuracy (PA)

PA is another measure of accuracy that can be computed using equation (3.24) (Elbayoumi et al., 2014).

$$PA = \frac{\sum_{i=1}^n (P_i - \bar{P})^2}{\sum_{i=1}^n (O_i - \bar{O})^2} \quad (3.24)$$

where  $n$  is the total number of sample,  $P_i$  is the predicted  $O_3$  concentration,  $\bar{P}$  is the mean of the predicted  $O_3$  concentration,  $O_i$  is the observed  $O_3$  concentration, and  $\bar{O}$  is the mean of the observed  $O_3$  concentration.

### Coefficient of determination ( $R^2$ )

$R^2$  indicates variability in the predicted data that can be explained by the fact that they are related to the observed values (Elbayoumi et al., 2014). The equation for  $R^2$  is as follows (Ul-Saufie et al., 2013):

$$R^2 = \left( \frac{\sum_{i=1}^n (P_i - \bar{P})(O_i - \bar{O})}{n \cdot S_{pred} \cdot S_{obs}} \right)^2 \quad (3.25)$$

where  $n$  is the total number of sample,  $P_i$  is the predicted  $O_3$  concentration,  $O_i$  is the observed  $O_3$  concentration,  $\bar{P}$  is the mean of the predicted  $O_3$  concentration,  $\bar{O}$  is the mean of the observed  $O_3$  concentration,  $S_{pred}$  is the standard deviation of the predicted  $O_3$  concentration, and  $S_{obs}$  is the standard deviation of the  $O_3$  concentration between input and output vectors.

## RESULTS AND DISCUSSIONS

### Descriptive Statistics of Ozone Concentration

Descriptive statistics of O<sub>3</sub> concentration at all stations is illustrated using box and whisker plot in Figure 9. The results showed that the whisker line that was connected to the minimum value was relatively shorter than whisker line connected to the maximum value. It indicated that O<sub>3</sub> concentrations in all stations were positively skewed or 'skewed to the right' and signifying the occurrences of extreme event. From the analysis of box plot, there were occurrences of extreme value at all sites. For stations such as Shah Alam, Kajang, Gombak, Ipoh and Seberang Jaya, the median is much lower as compared to the mean and showed large differences in these values. Even, the other stations showed smaller differences, but still, it is indicate that there are an extreme event occurs throughout 12 years of study period.

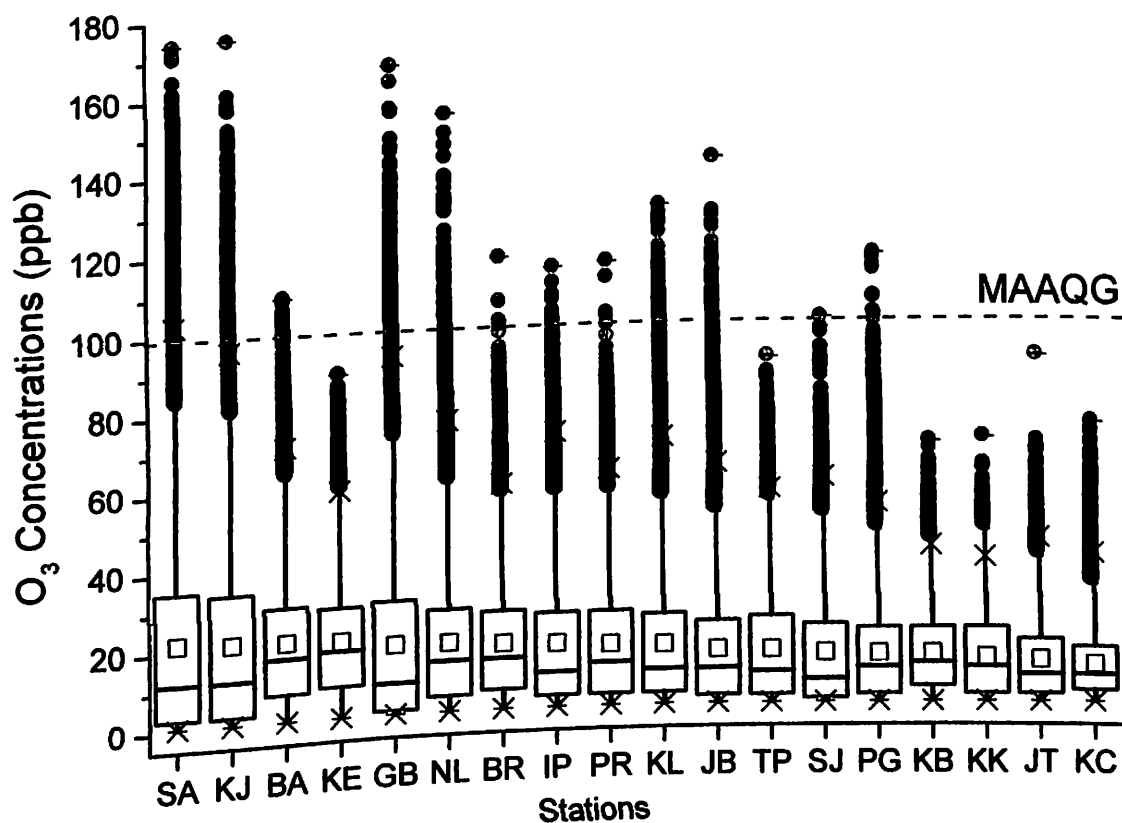


Figure 9 Box and whisker plot of hourly average O<sub>3</sub> concentration for all stations

Relatively high O<sub>3</sub> concentration observed at stations located in urban and industrial areas. The highest concentration was recorded in Kajang with 175 ppb followed by Shah Alam with 174 ppb. Maximum concentration in most of the stations except for Kemaman, Kota Bharu, Kota Kinabalu, Jerantut and Kuching were higher than 100 ppb which surpass the recommended MAAQG value of 100 ppb. Whereas, minimum O<sub>3</sub> concentrations were consistently measured at 1 ppb for all stations.

## **OZONE FLUCTUATIONAL BEHAVIOUR**

Ground level O<sub>3</sub> exhibited strong fluctuation behaviour due to changes in precursor concentrations, photochemical reactions and weather conditions. Fluctuational behaviour is very important in order to understand the O<sub>3</sub> characteristics in each study areas.

### **Daily Fluctuations**

The time series plots of daily maximum O<sub>3</sub> concentrations for 12 years are illustrated in Figure 10 to Figure 12. The reference line showed the MAAQG for one-hour averaging time of 100 ppb were drawn in the plots to show the exceedances that occurred during the study period. Results showed that O<sub>3</sub> concentrations fluctuated throughout the study period with significant higher concentration observed at several urban and industrial stations such as Shah Alam, Kajang, Gombak, Klang, Johor Bahru and Nilai. The maximum daily O<sub>3</sub> concentrations at these stations ranged from 129 ppb to 175 ppb. Similar findings have been discussed in several studies (Ghazali et al., 2010; Latif et al., 2011; Banan et al., 2013; Ahamad et al., 2014) which stated that high O<sub>3</sub> concentrations were recorded in urban and industrial areas.

Most of these stations are located in Klang Valley which is one of the most heavily populated area in Malaysia and centre of various economic activities. High density residential areas and multilane highways are constructed to accommodate high population density. Over this area, various anthropogenic activities become major sources of O<sub>3</sub> precursors and contributed to elevate the O<sub>3</sub> concentration under



suitable weather conditions. The findings are in line with the previous studies which identified traffic and industrial emissions as the most eminent sources of O<sub>3</sub> precursors in Klang Valley (Ishii et al., 2007; Ghazali et al., 2010; Latif et al., 2012). High population and traffic density also contribute to congestion especially during morning and late afternoon rush hour. Low speed vehicles release higher concentration of NO (Banan et al., 2013) and subsequently elevates NO<sub>2</sub> production which is the most important O<sub>3</sub> precursor (Tiwary and Colls, 2009).

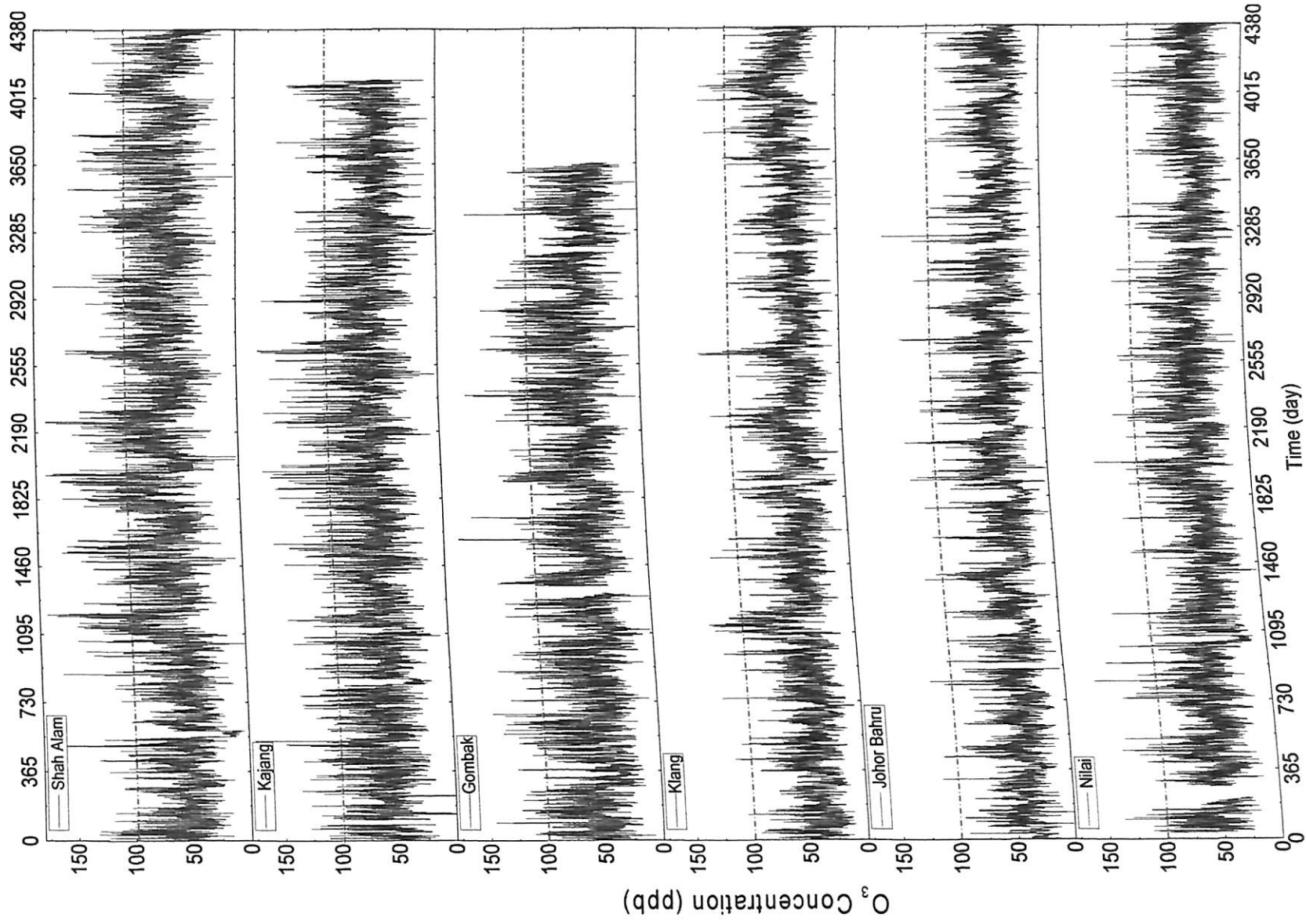


Figure 10 Time series plot of daily maximum O<sub>3</sub> concentration for Shah Alam, Kajang, Gombak, Klang, Johor Bahru and Nilai

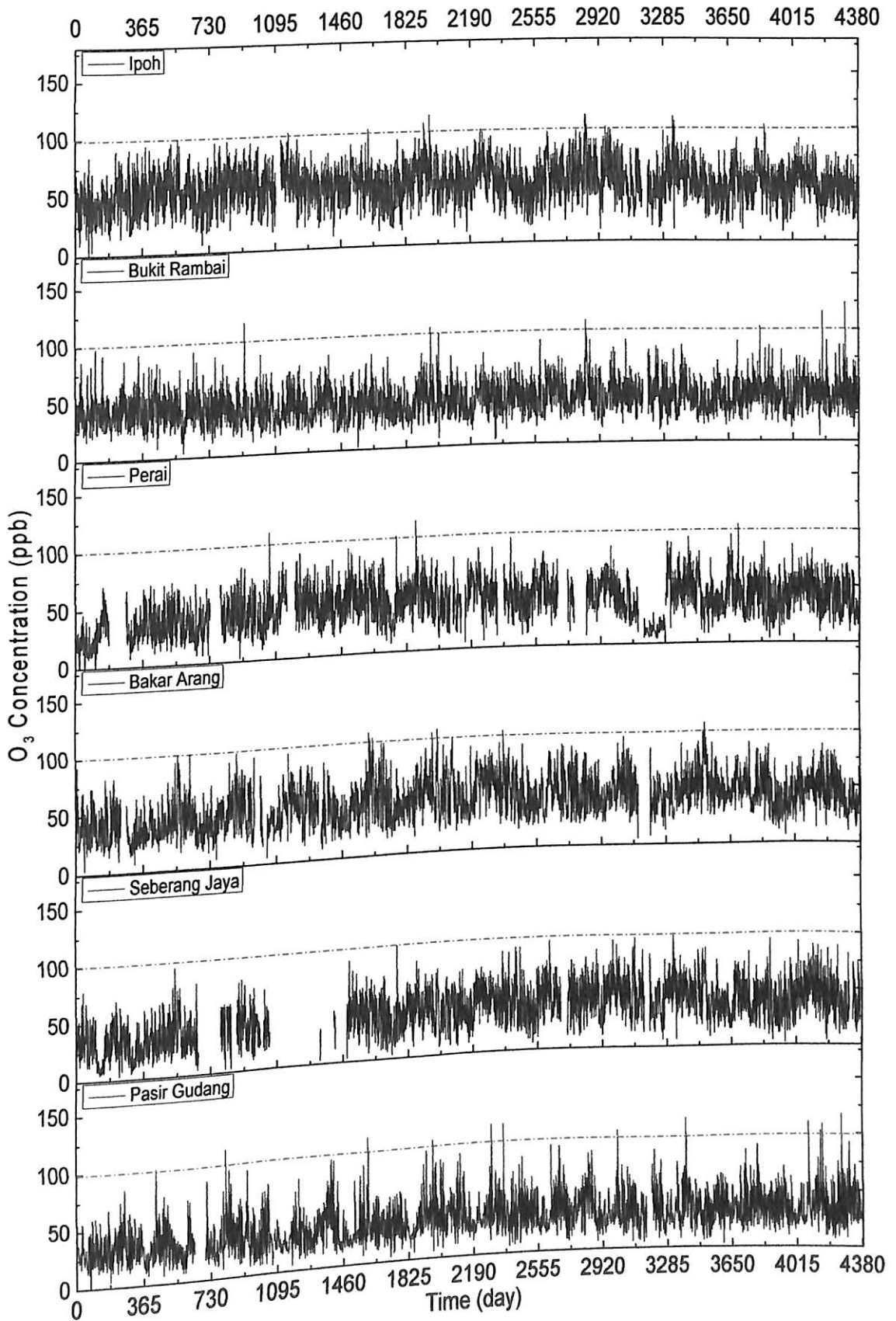


Figure 11 Time series plot of daily maximum O<sub>3</sub> concentration for Ipoh, Bukit Rambai, Perai, Bakar Arang, Seberang Jaya, Pasir Gudang

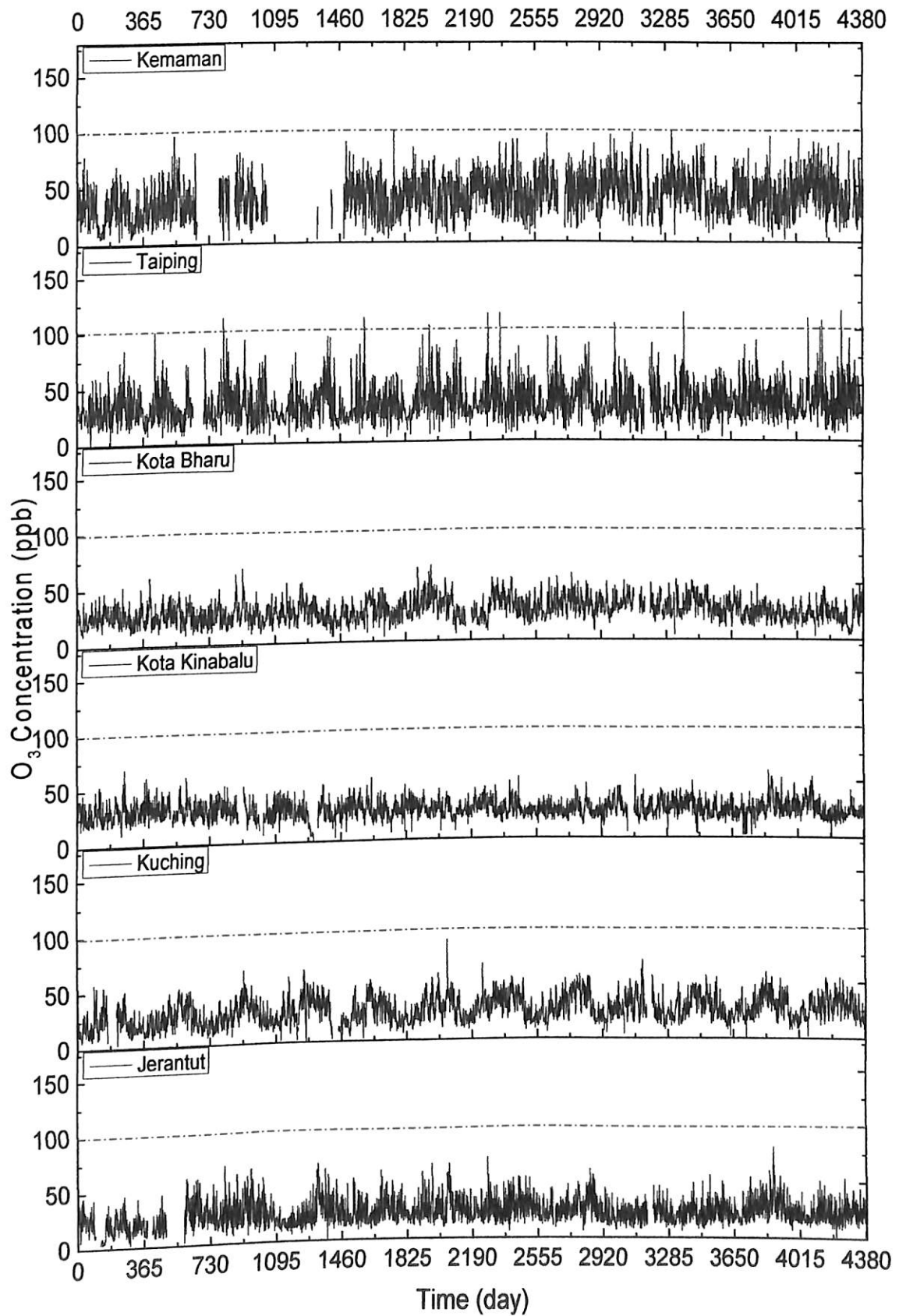


Figure 12 Time series plot of daily maximum O<sub>3</sub> concentration for Kemaman, Taiping, Kota Bharu, Kota Kinabalu, Kuching, Jerantut

## Diurnal Fluctuations

Since solar radiation plays the most significant role in  $O_3$  production, diurnal variations have a major influence on the  $O_3$  formation and concentration in the atmosphere. Diurnal plots of hourly average  $O_3$  concentrations is depicted in Figure 13. Typical diurnal trends characterized by high concentration during DT and low concentration during NT were observed at all stations. Similar diurnal pattern of  $O_3$  concentration during NT were observed at all stations. Similar diurnal pattern of  $O_3$  concentration were observed in numerous areas around the world (Zhang and Kim Oanh., 2002; Duenas et al., 2004; Han et al., 2011; Reddy et al., 2011). Influence of the incoming solar radiation in  $O_3$  photochemical production contributed to the findings. It is established that  $O_3$  is only formed during DT (Seinfeld and Pandis, 2006; Tiwary and Colls, 2009). During NT,  $O_3$  is destructed through several pathways that involved reactions with NO (NO titration) or with  $NO_2$  to yield  $NO_3$  (Ghosh et al., 2013).

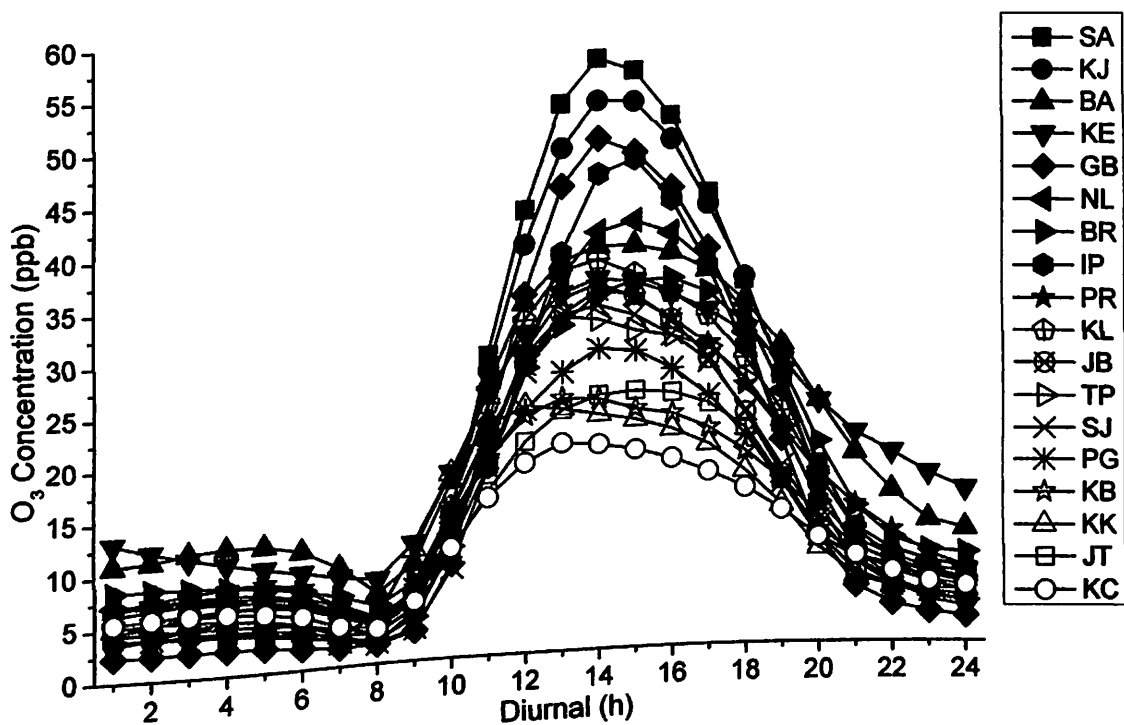


Figure 13 Diurnal plot of hourly average  $O_3$  concentrations in all stations

Malaysia starts to receive sunlight approximately at 7 a.m. daily. With the availability of sunlight and precursors,  $O_3$  starts to be produced by photochemical reactions. However,  $O_3$  reached their minimum concentrations at 8 a.m. and this

scenario was observed in all stations except for Gombak and Kota Kinabalu. The minimum concentrations was measured at 7 a.m. for Gombak and Kota Kinabalu stations due to NO titrations. At most of the stations, the maximum O<sub>3</sub> was reached at 2 p.m.. Nilai, Ipoh and Jerantut reached the maximum O<sub>3</sub> at 3 p.m. while Taiping, Kota Bharu and Kuching at 1 p.m.. The differences in the maximum concentration time was expected due to the time of incoming solar radiation. Kota Kinabalu and Kuching which are located in the East of Malaysia received earlier sunlight compared to the Peninsular Malaysia, hence contribute to earlier photochemical reactions of the day.

Local precursor's emission is also an important influencing factor. Wang et al. (2008) claimed that if the precursors produced remained localized, the peak of O<sub>3</sub> concentrations usually occur around noon. However, if the precursors are being transported, the peak will be delayed and will occur in the late afternoon. Besides the effects of solar radiation and variation in precursor sources, O<sub>3</sub> photochemical reactions were also being influenced by boundary layer processes and meteorological parameters such as wind and relative humidity. According to Ghazali et al. (2010), these parameters also play important roles in O<sub>3</sub> diurnal variations.

During late afternoon and evening, O<sub>3</sub> concentrations decreased due to the reduction in O<sub>3</sub> production efficiency and gradually decreased during NT due to several destruction mechanisms such as transformation, deposition and transportation. The results suggested that there are large differences between DT and NT O<sub>3</sub> concentrations.

### **Spatial fluctuations**

GIS analysis was used in order to illustrate the spatial fluctuation of ground level O<sub>3</sub> graphically. Blond et al. (2007) claimed that the pollutant concentration map was a very useful method to deliver information to citizens about the quality of the air that they are breathing regardless of whether or not they live close to the monitoring stations. The worst case scenario of O<sub>3</sub> pollution in Malaysia can be illustrated by the annual maximum O<sub>3</sub> map in Figure 14 as the map was developed using the highest hourly O<sub>3</sub> concentrations for each year during 1999 to 2010. The results



clearly indicated that more serious O<sub>3</sub> pollutant problems were observed in the west coast of Peninsular Malaysia that included Selangor and Negeri Sembilan.

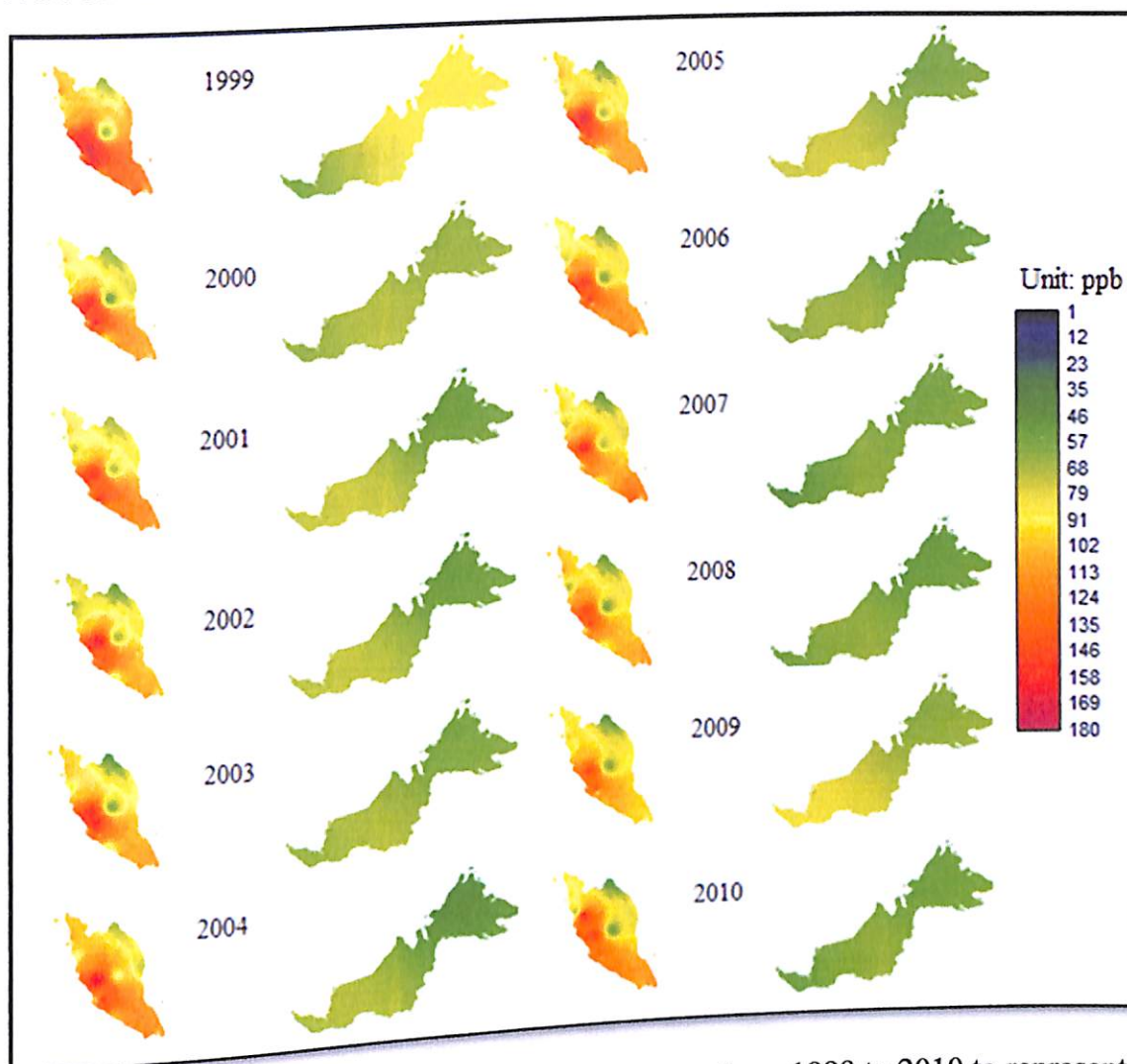


Figure 14 Annual maximum O<sub>3</sub> concentration map from 1999 to 2010 to represent the worst case scenario (unit in ppb)

Southern states such as Johor and Malacca were also affected by O<sub>3</sub> pollutant at a lower degree than the middle of west coast. Ipoh became the most pronounced area affected by O<sub>3</sub> pollution in the northern regions. However, in 2010 Bakar Arang emerged to be one of the highly affected areas in Peninsular Malaysia. The map also exhibited that the east coast of Peninsular Malaysia (Kelantan and Terengganu), middle of Pahang, Sabah and Sarawak were less impacted by the O<sub>3</sub> pollution. These states can be the cleanest areas in Malaysia from O<sub>3</sub> pollutants. Significantly low O<sub>3</sub> concentrations were observed in Jerantut over 1999 to 2010 due to the location of this station in rural areas close to the reserve forest. According to Latif et al. (2014), air quality in Jerantut is important to evaluate air quality status pre and

post modernisation. The fluctuation behaviour of the O<sub>3</sub> concentrations can be detected from the map which clearly indicated that O<sub>3</sub> concentration was relatively higher during 1999 and slowly decreased through the study period, with the most affected areas in the west coast of Peninsular Malaysia. However, in 2010, the most affected areas were shifted which made the most affected areas located in the northern region of Peninsular Malaysia.

### Exceedance Analysis of Hourly O<sub>3</sub> Concentrations

The exceedance analysis of hourly O<sub>3</sub> concentration was carried out in order to investigate the numbers, time of occurrences, and location, which the exceedances happened. Ahamad et al. (2014) expressed that identification of stations that frequently showed exceedance cases is important when formulating abatement and mitigation plans. One exceedance is defined when 1-hour average of O<sub>3</sub> concentrations was over the MAAQG limit of 1-hour average of 100 ppb. The number of exceedance based on the occurrence years is depicted in Figure 15.

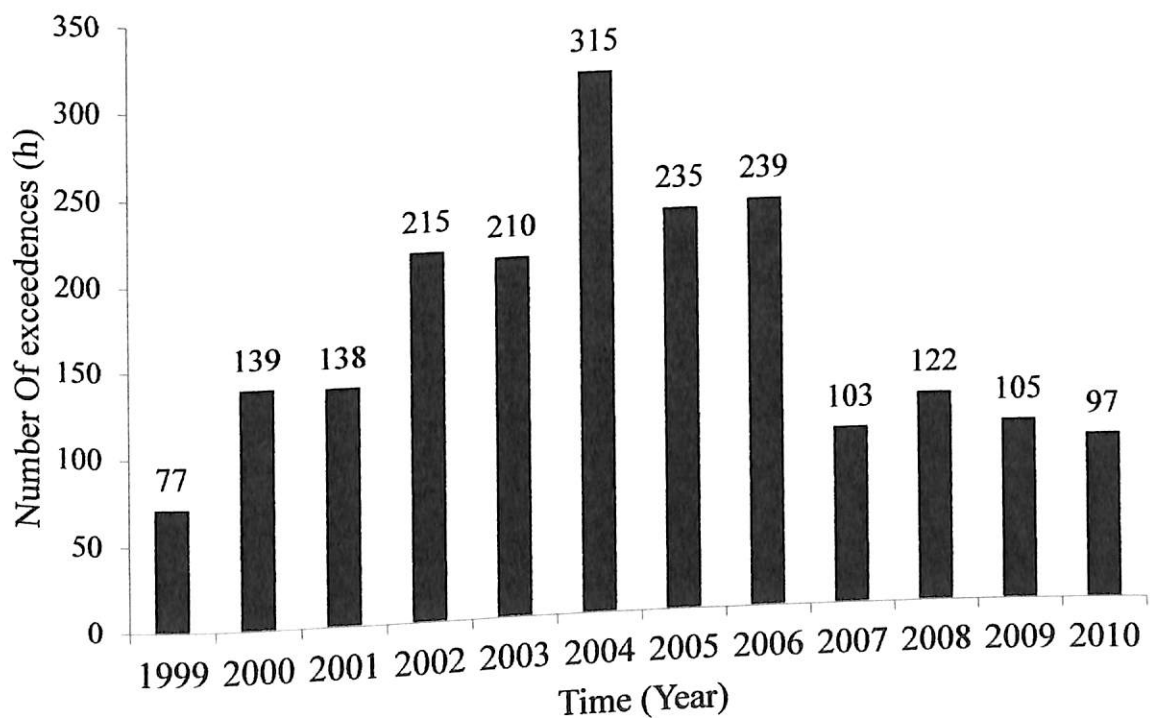


Figure 15 Annual number of exceedances in all stations

The total number of exceedances in all stations during 1999 to 2010 was 1995 hours or 166.25 hour/year average number of exceedance. The results demonstrated that



the highest number of exceedance was recorded in 2004 with 315 hours, while the lowest exceedance was in 1999 with 77 hours. High number of exceedances was also recorded in 2002, 2003, 2005 and 2006 with 215 hours, 210 hours, 235 hours and 239 hours, respectively. High number of exceedances during 2002, 2003, 2004, 2005 and 2006 coincided with serious high particulate events in Southeast Asia (Tangang et al., 2010). Heil and Goldammer (2001) alleged that Southeast Asia pollutants episodes outbreaks were mostly due to biomass burning that release large variety of pollutants such as PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO, CO<sub>2</sub> and hydrocarbons. Surged in precursors concentrations during the episodes will eventually increase O<sub>3</sub> concentration, thus lead to high number of exceedances during the period.

Result exhibited that the number of exceedances was significantly lower from 2007 to 2010 even the number of registered motor vehicles in Malaysia is in increasing trend as showed in Figure 7. The scenario may contributed by implementation of new emission standard for light duty vehicles in 2009, which upgraded from Euro 1 to Euro 2M. Implementation of Euro 2M standards tremendously reduced number of sulphur emission from 2500 ppm (Euro 1) to 500 ppm (Euro 2M), thus lead to reduction of NO<sub>x</sub> emission by motor vehicles.

Fluctuations in exceedances occurrence can also be contributed by the locations where the stations are established. The number of exceedances based on location and type of monitoring station is illustrated in Figure 16. The plot illustrated that most numbers of exceedances were recorded in the urban stations with 1865 hours or 93.5% of total exceedances followed by the industrial stations (120 hours, 6%) and sub urban stations (10 hours, 0.5%). Meanwhile, zero exceedance was observed in the background station.

These observations showed that the O<sub>3</sub> exceedances pattern is highly depended on localised pollutant emissions. High chances of O<sub>3</sub> concentrations exceed the guideline limits at urban areas since the sources of O<sub>3</sub> precursors are abundant. In the city or urban areas, the wind is often obstructed by manmade buildings that cause the wind speed to slower down and may initiate O<sub>3</sub> pollution accumulations (Chan et al., 2001). Kim and Guldmann (2011) also mentioned that the pollutants associated with traffic were at highest ambient concentration levels when wind speed was low.

The findings is in line with the study by Ahamad et al. (2014) that claimed that the characteristic features of the pollutant emissions and mixing at each stations appear to distinguish the stations that consistently showed high or low exceedances.

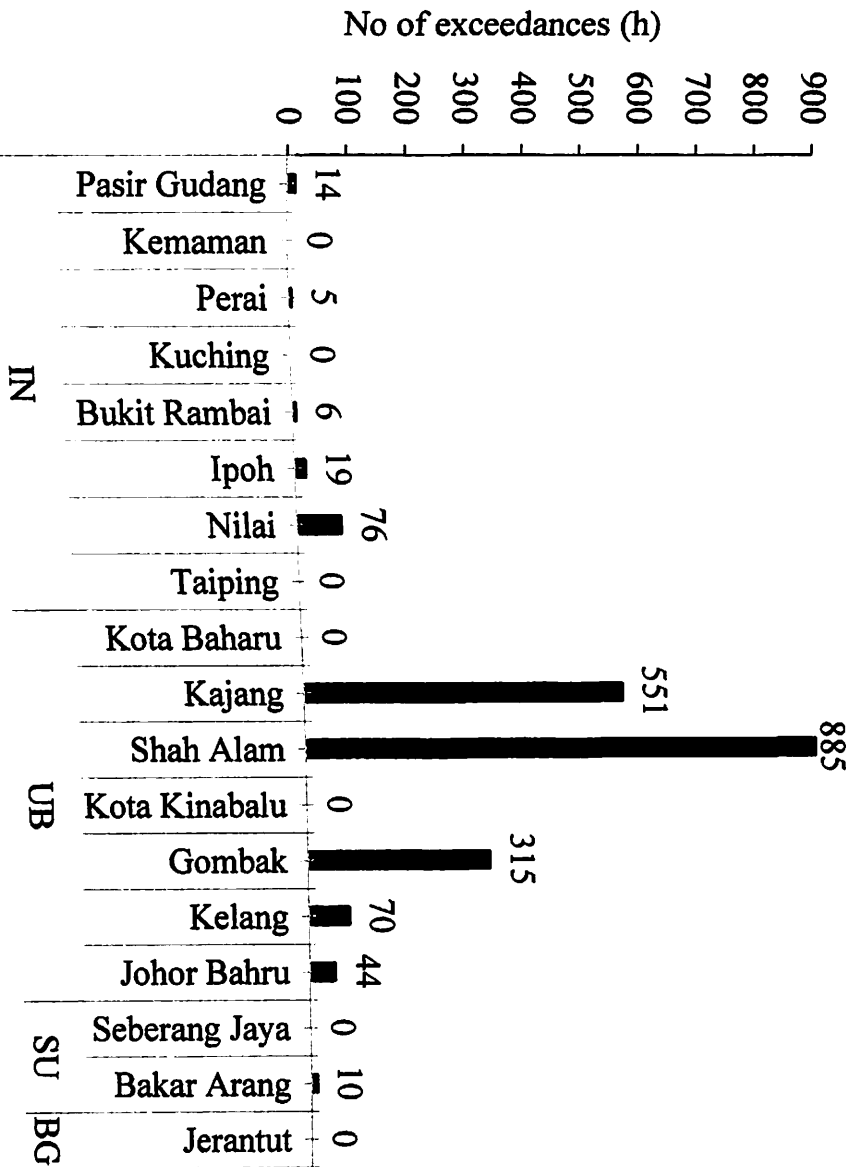


Figure 16 Numbers of exceedances in all stations

Numbers of exceedances of O<sub>3</sub> concentration based on diurnal variation was also explored and the results are illustrated in Figure 17. The plot pattern suggested that the number of exceedances followed the typical O<sub>3</sub> diurnal plot. Most of exceedances were recorded at 3 p.m. with 600 hours. The earliest time the recorded exceedances occurrence was at 10 a.m., while the latest exceedance measured was at 6 p.m.. Since solar radiation was the predetermined factor in O<sub>3</sub> production, the increase in solar radiation would contribute to the increase of O<sub>3</sub> concentration.

Malaysia received constant solar radiation from 7 a.m. to 7 p.m. daily (Mohammed et al., 2013). Production of O<sub>3</sub> is expected to start, as the solar radiation is available and have been accumulated to a higher concentration with the increase of the O<sub>3</sub> precursor and radiation intensity. Diurnal profile of O<sub>3</sub> concentration in Figure 5

showed that  $O_3$  will start to accumulate from 9 a.m.. Due to high concentrations of their precursors,  $O_3$  was able to accumulate at a higher rate, thus lead to exceedances as early as 10 a.m.. Nevertheless, the highest number of exceedances were measured at 3 p.m., an hour later than the maximum time of  $O_3$  concentrations which usually observed at 2 p.m..

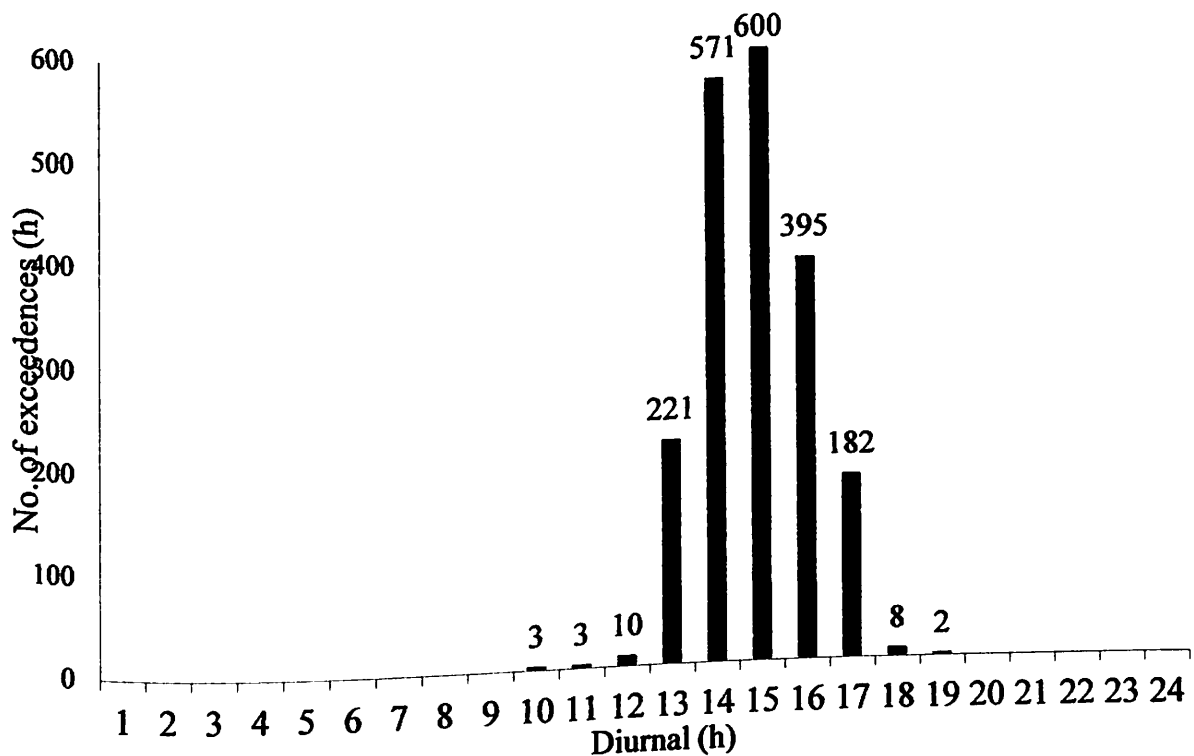


Figure 17 Diurnal variations of  $O_3$  exceedances

### Ozone Transformational Behaviour

Ozone transformational behaviours varied according to changes in concentration of  $NO_2$  and  $NO$  as well as temperature and incoming solar radiation. Due to the effect of the precursors and meteorological parameters,  $O_3$  showed distinctive characteristics during morning. The critical conversion points (CCP) which are assumed as a starting point when  $NO_2$  photolysis rate began to be higher than  $NO$  titration rate is determined based on these variables. CCP is important because  $O_3$  starts to accumulate only after the CCP.

### Influence of NO<sub>2</sub>, NO, Temperature and UVB to O<sub>3</sub> Formation

At ground level, the O<sub>3</sub> production is controlled by their precursor's concentration and incoming solar radiation intensity. In this study, PCA is used to statistically determine the contributions of NO<sub>2</sub>, NO, T and UVB as well as to select the biggest contributor to formations of ground level O<sub>3</sub> among these important precursors and meteorological factors.

The PCA results for all stations are shown in Table 7. From the percentage of the total variance explained, PCA established that NO<sub>2</sub>, NO, T and UVB were able to explain variations in O<sub>3</sub> concentrations up to 81.50% (Kota Bharu).

Table 7 Principal component analysis on NO, NO<sub>2</sub>, temperature and UVB

Stations	PC	NO	NO <sub>2</sub>	T	UVB	E	V	ZV	Group
SA	PC1	0.794	0.869	-0.015	-0.283	1.466	36.65	72.65	P
	PC2	-0.212	-0.055	0.889	0.776	1.440	36.00		M
KJ	PC1	-0.172	-0.042	0.916	0.826	1.553	38.83	71.94	M
	PC2	0.728	0.841	0.010	-0.294	1.324	33.11		P
BA	PC1	-0.048	-0.294	0.885	0.884	1.653	41.34	78.34	M
	PC2	0.895	0.802	-0.132	-0.183	1.494	37.34		P
KE	PC1	-0.012	-0.027	0.798	0.767	1.226	30.66	58.70	M
	PC2	0.685	0.781	0.118	-0.165	1.122	28.04		P
GB	PC1	-0.449	0.168	0.825	0.792	1.537	38.42	68.21	M
	PC2	0.614	0.863	0.179	-0.193	1.191	29.79		P
NL	PC1	-0.563	-0.686	0.850	0.868	2.263	56.57	56.57	-
BR	PC1	0.847	0.847	0.058	-0.176	1.469	36.73	68.64	P
	PC2	-0.024	-0.092	0.822	0.769	1.276	31.91		M
IP	PC1	0.823	0.852	0.159	-0.229	1.482	37.04	68.50	P
	PC2	-0.047	-0.007	0.819	0.765	1.258	31.46		M
PR	PC1	0.826	0.850	0.013	-0.199	1.444	36.11	71.21	P
	PC2	-0.115	-0.058	0.862	0.803	1.404	35.10		M
KL	PC1	0.850	0.878	0.050	-0.214	1.542	38.54	74.09	P
	PC2	-0.102	-0.047	0.870	0.808	1.422	35.55		M
JB	PC1	0.825	0.872	0.033	-0.350	1.566	39.14	71.90	P

	PC2	-0.114	-0.115	<b>0.891</b>	<b>0.701</b>	1.310	32.76		M
TP	PC1	<b>0.776</b>	<b>0.863</b>	0.175	-0.277	1.455	36.39	65.65	P
	PC2	-0.225	0.134	<b>0.846</b>	<b>0.621</b>	1.170	29.26		M
SJ	PC1	0.100	-0.017	<b>0.826</b>	<b>0.845</b>	1.742	43.56	68.61	M
	PC2	<b>-0.588</b>	<b>0.994</b>	0.041	0.042	1.002	25.06		P
PG	PC1	<b>0.814</b>	<b>0.852</b>	0.045	-0.255	1.455	36.39	70.60	P
	PC2	-0.133	-0.046	<b>0.869</b>	<b>0.771</b>	1.369	34.21		M
KB	PC1	-0.037	-0.136	<b>0.936</b>	<b>0.914</b>	1.732	43.31	81.50	M
	PC2	<b>0.864</b>	<b>0.862</b>	0.002	-0.197	1.528	38.19		P
KK	PC1	<b>0.866</b>	<b>0.829</b>	-0.052	-0.175	1.469	36.73	73.03	P
	PC2	-0.039	-0.193	<b>0.854</b>	<b>0.827</b>	1.452	36.30		M
JT	PC1	0.046	-0.074	<b>0.808</b>	<b>0.818</b>	1.329	33.24	59.90	M
	PC2	<b>0.416</b>	<b>0.878</b>	0.267	-0.227	1.067	26.67		P
KC	PC1	0.043	-0.055	<b>0.877</b>	<b>0.880</b>	1.549	38.72	76.46	M
	PC2	<b>0.866</b>	<b>0.869</b>	0.037	-0.050	1.510	37.74		P

Note: E is eigenvalue; V is variance;  $\Sigma V$  is cumulative variance; P is precursor; M is meteorology parameters.

In overall, the total cumulative variances are in range of 60 to 80% except for Kemaman, Nilai and Jerantut which are 58.70%, 56.57% and 59.90%, respectively. The findings proved that NO<sub>2</sub>, NO, T and UVB are the most significant factors that controlled the O<sub>3</sub> production and variation in lower atmosphere due to the ability of these variables to explain the O<sub>3</sub> variations up to 80%.

The results demonstrated that the influence of these variables were significantly higher in urban areas as the percentage of total variances in Shah Alam, Kajang, Klang, Johor Bahru, Kota Bharu and Kota Kinabalu was 72.65%, 71.94%, 74.09%, 71.90%, 81.50% and 73.03%, respectively. Whereas, the percentage was slightly lower in industrial areas such as Kemaman, Bukit Rambai, Ipoh and Taiping which recorded 58.70%, 68.64%, 68.50% and 65.65%, respectively. Low percentage of variation was measured for Jerantut with 59.90%. However, the results was expected, since the station served as the background stations which the anthropogenic sources that were considered minimal.

PCA results clearly separated  $\text{NO}_2$  and  $\text{NO}$  concentration from T and UVB into two separate PCs except in Nilai. However, no consistency of either the precursors or the meteorological parameters were selected as PC1 or PC2. Basically in PCA, the highest percentage of variance is denoted as PC1, and lower percentage of variance is called PC2. At nine stations (Shah Alam, Bukit Rambai, Ipoh, Perai, Klang, Johor Bahru, Taiping, Pasir Gudang and Kota Kinabalu), PCA selected precursors ( $\text{NO}_2$  and  $\text{NO}$ ) as PC1 while at other stations meteorological parameters (T and UVB) are the PC1. On a contrary, PCA results in Nilai only produced one PC with the total variance explained of 56.57%.

Result demonstrated that at stations where precursors is dominant factor (PC1) such as Shah Alam, Ipoh and Perai, high traffic density and frequent congestion is observed. Evaporation losses from the fuel tank and carburettor combined with pollutants emitted from exhaust gas, are the major factors causing air pollution by motor vehicles. In Malaysia, Zainordin et al. (2014) claimed that motor vehicle emissions are one of the major sources of urban air pollutants such as PM,  $\text{NO}_2$ ,  $\text{SO}_2$ , CO, Pb, benzene and VOCs. Any disturbance in traffic such as change in free flow traffic to intersection can lead to changes in speed cycle, which in turn increases the mean delay and queue length and eventually cause an increment in emissions rates (Zainordin et al. 2014).

The influence of  $\text{NO}_2$ ,  $\text{NO}$ , T and UVB were also studied using the composite diurnal plot of  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{NO}$ , T and UVB as shown in Figure 10 to Figure 15. Diurnal variations have significant influences towards  $\text{O}_3$  formation and fluctuation behaviour in the atmosphere. The physical and chemical characteristics of  $\text{O}_3$  concentrations at a certain location can be analysed based on diurnal behavioural characteristics (Reddy et al., 2011).  $\text{O}_3$  demonstrates similar diurnal trends with T and UVB and inverse trends was observed between  $\text{O}_3$  and  $\text{NO}_x$ .  $\text{O}_3$ , T and UVB showed uni-modal variations with a single peak values around noontime, while  $\text{NO}_2$  and  $\text{NO}$  exhibited double peaks. This trend was consistently observed in all stations and the obtained results are in line with various studies worldwide (Duenas et al., 2004; Reddy et al., 2011; Han et al., 2012; Ahamad et al., 2014; Iqbal et al., 2014). During daytime, high  $\text{O}_3$  concentration, T and UVB was observed, while during nighttime  $\text{NO}_2$  and  $\text{NO}$  concentrations were relatively higher than the other

parameters.  $O_3$  photochemical reactions only occur during daytime due to solar radiation factors and in all stations the solar radiation began after 7 a.m. and gradually increased during morning. During the period, daily  $O_3$  photochemical reactions also start. However, at 8 a.m.,  $O_3$  is at their minimum concentration due to scavenge by NO titrations (Tiwary and Colls, 2009; Jiménez-Hornero et al., 2010).

During morning rush hour, usually around 7 a.m. to 9 a.m., high concentrations of NO is emitted by large volume of vehicles. Maximum NO in all stations was constantly measured at either 8 a.m. or 9 a.m. except for Kota Kinabalu which at 7 a.m.. High NO concentrations were observed in several stations such as Gombak, Klang and Seberang Jaya. At these stations, the diurnal maximum NO concentrations were above 50 ppb. At the same period,  $NO_2$  concentration also showed an increasing trend due to the intense rate of NO titration which converted NO into  $NO_2$ .

The results exhibited that  $NO_2$  concentrations reach their morning peak concentrations mostly at 9 a.m. or 10 a.m., which is about an hour later than NO. Meanwhile, maximum UVB up to  $800 J/m^2h$  was constantly measured in between 1 p.m. to 2 p.m.. At the same period, the temperature also reached their daily maximum value in the range of 30 to 32 °C, while  $NO_2$  and NO concentrations showed the opposite trends. The formation of  $O_3$  is heavily influenced by sunlight and temperature (Barrero et al., 2006; Kovač-Andrić et al., 2009) as high UVB will favour  $O_3$  formation which is a photochemical species.

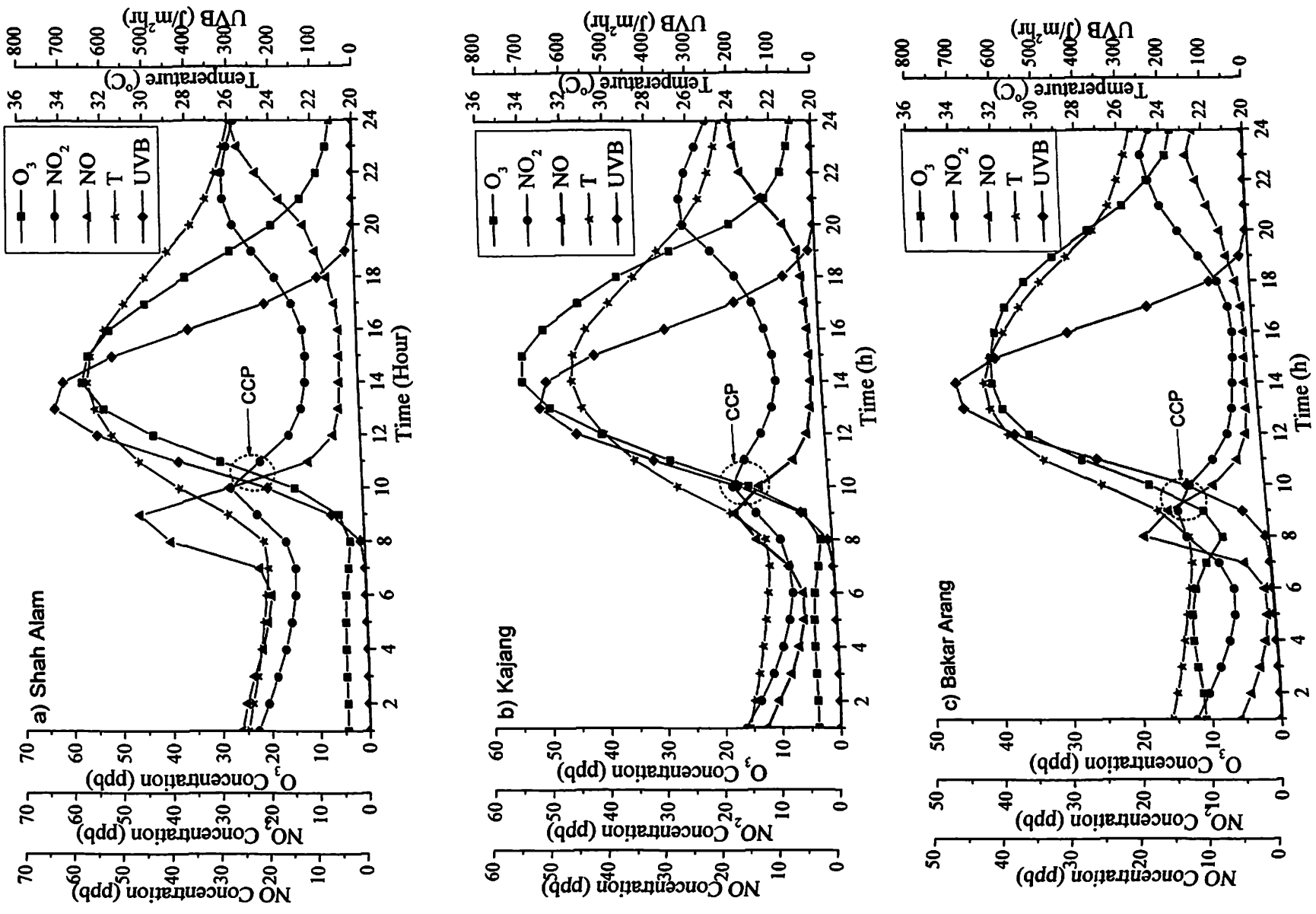


Figure 18 Composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, temperature and solar radiation in Shah Alam, Kajang and Bakar Arang



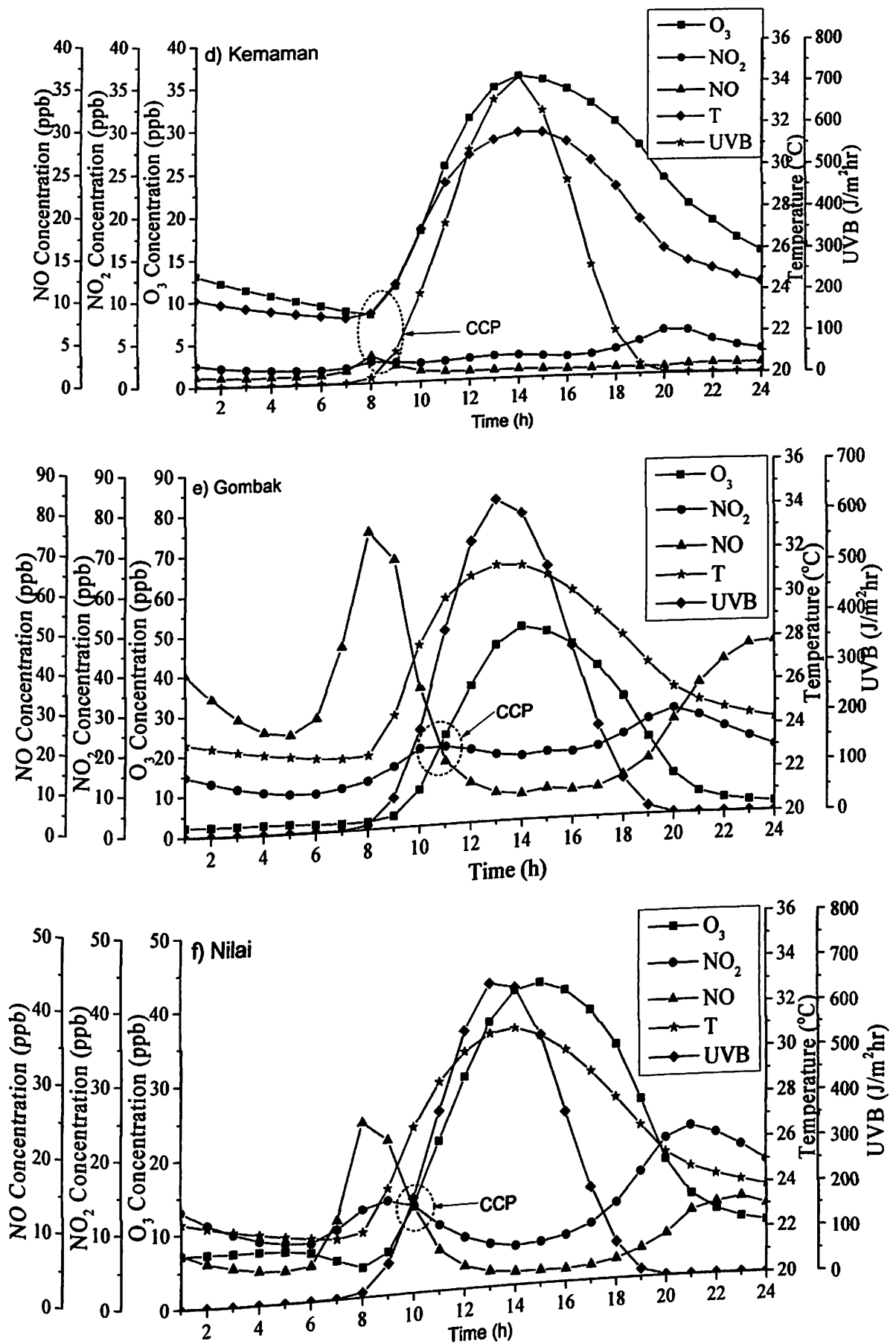


Figure 19 Composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, temperature and solar radiation in Kemaman, Gombak and Nilai

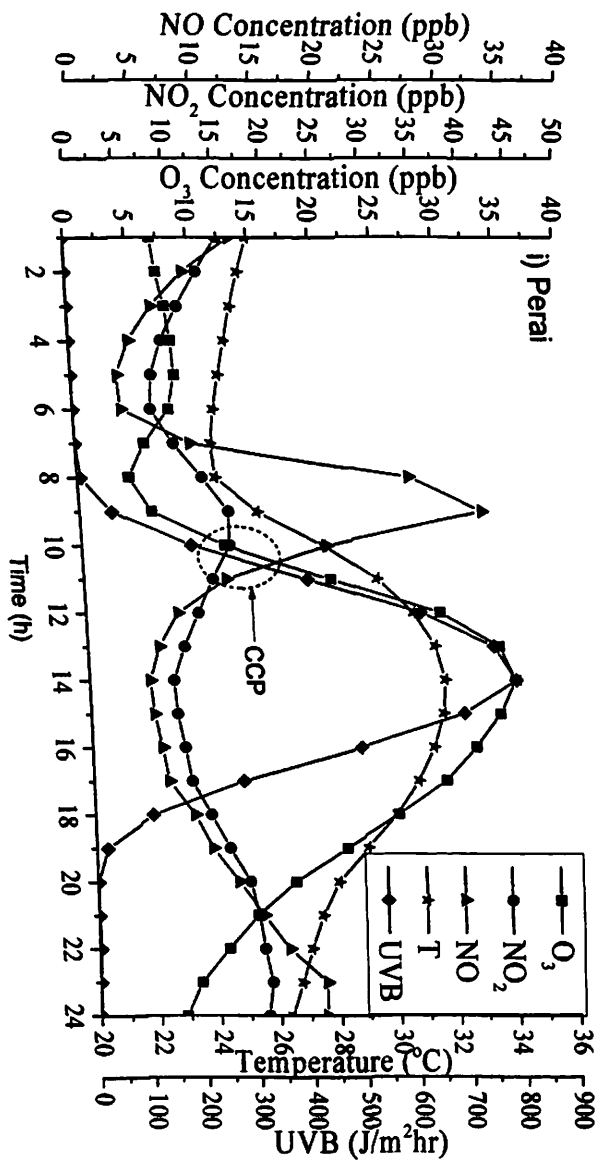
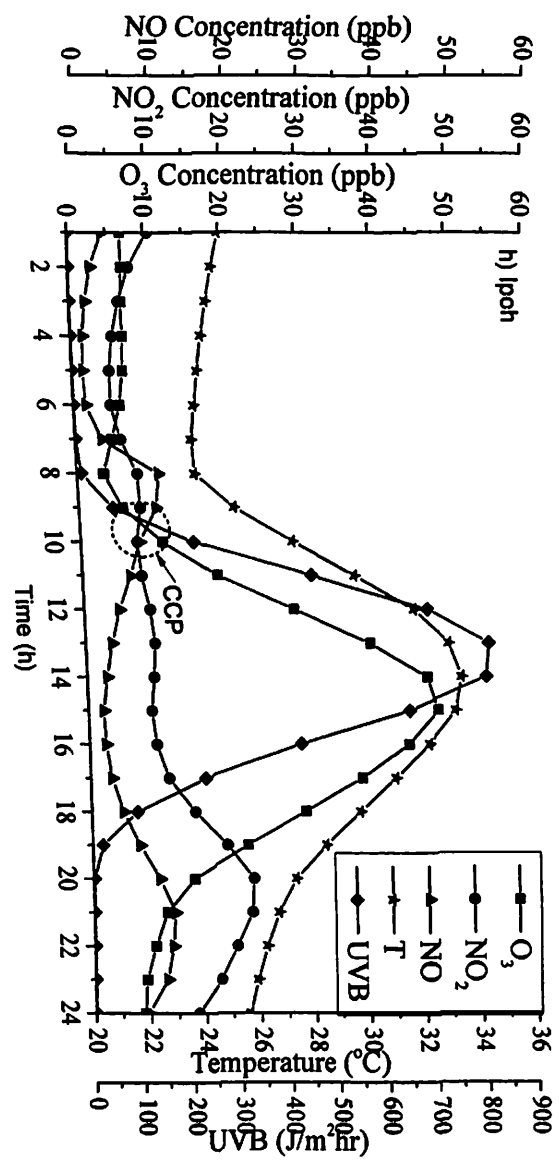
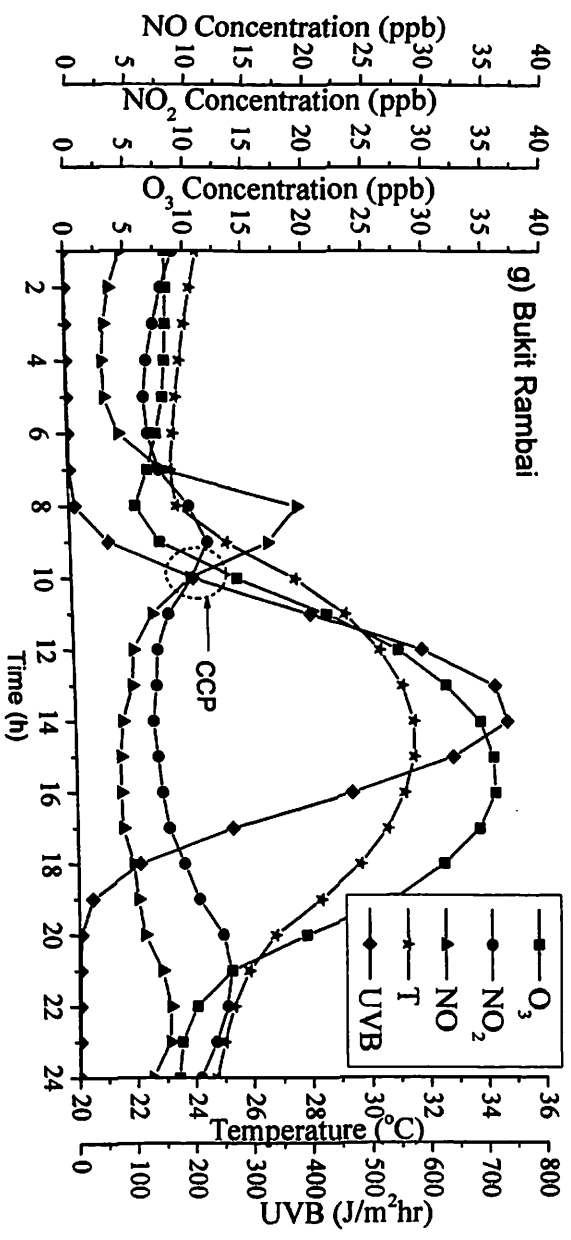


Figure 20 Composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, temperature and solar radiation in Bukit Rambai, Ipoh and Perai

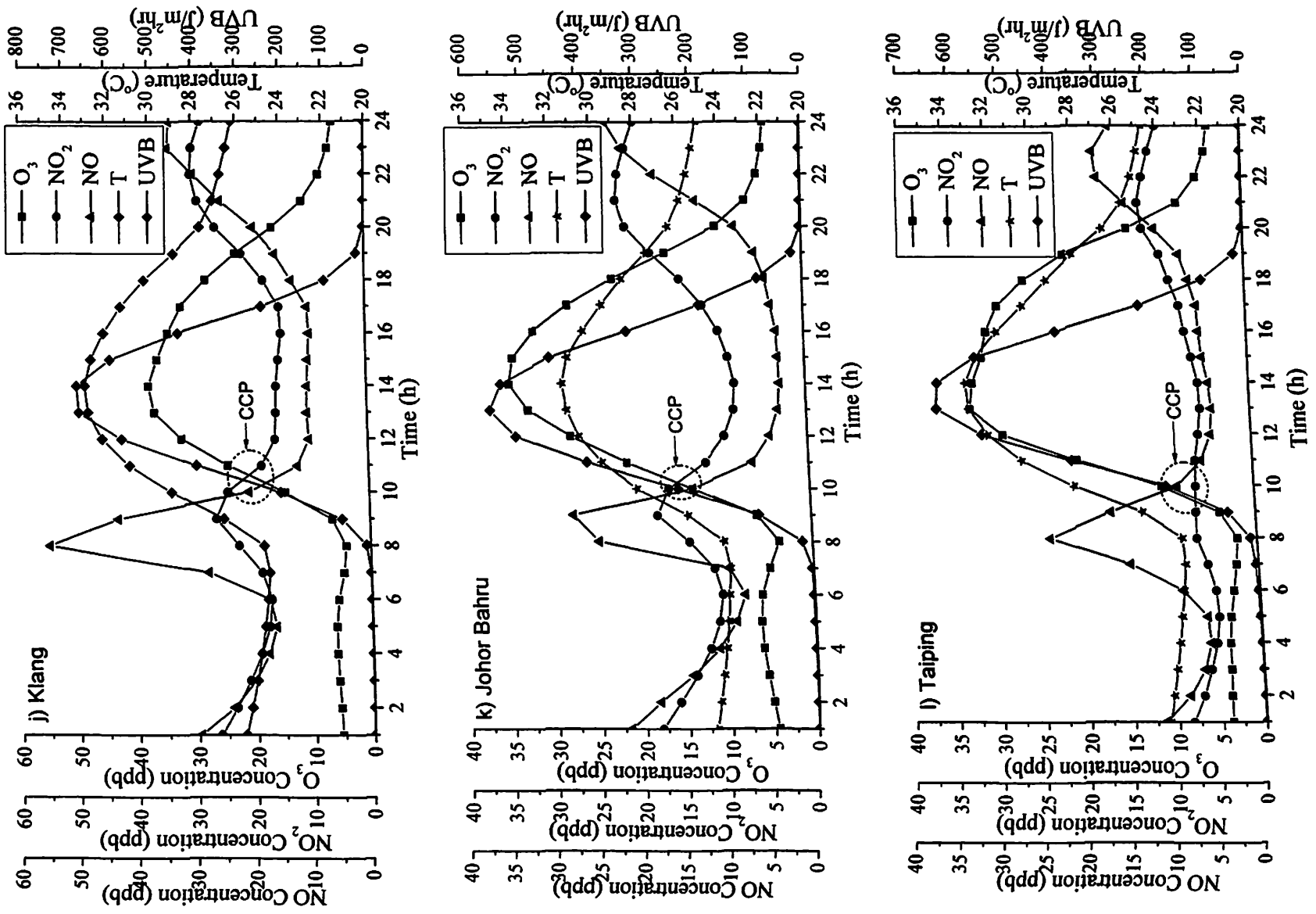
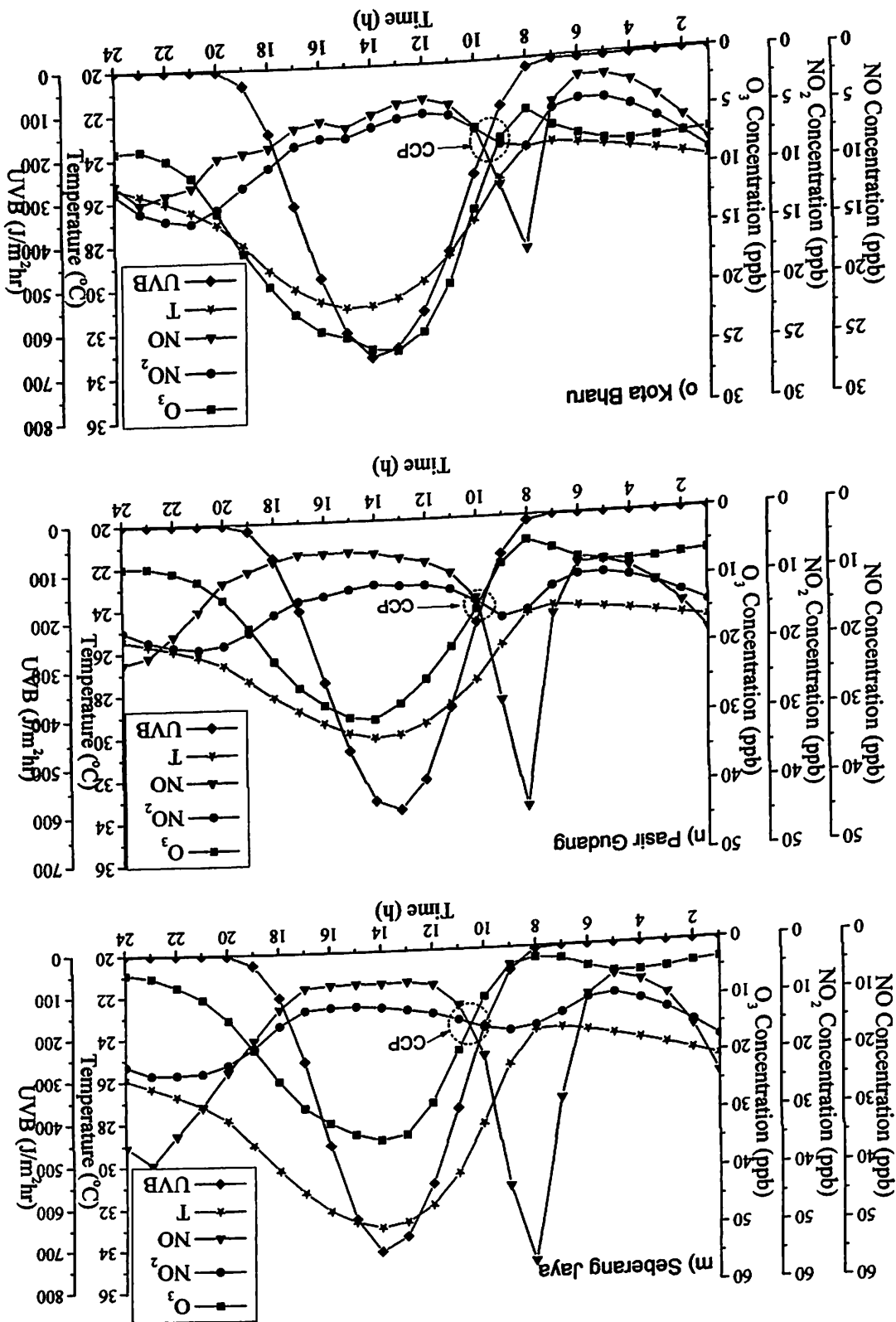


Figure 21 Composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, temperature and solar radiation in Klang, Johor Bahru and Taiping

Figure 22 Composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, temperature and solar radiation in Seberang Jaya, Pasir Gudang and Kota Bharu



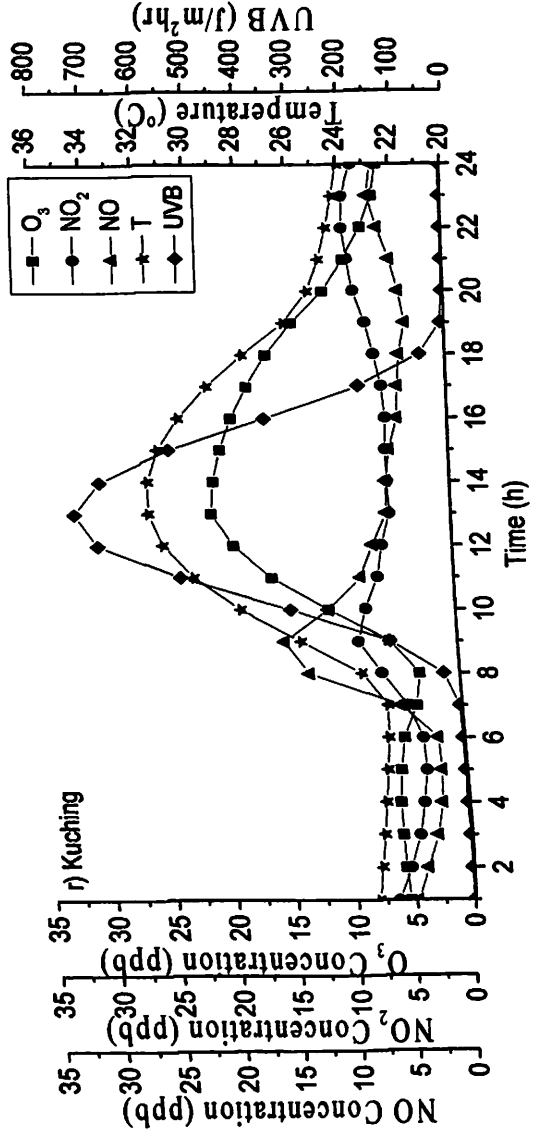
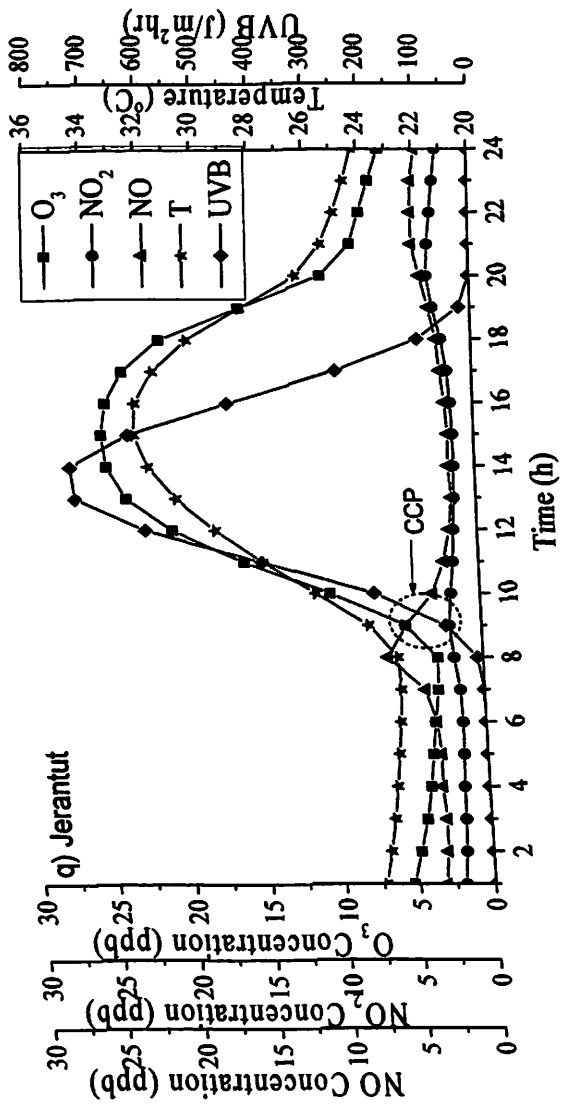
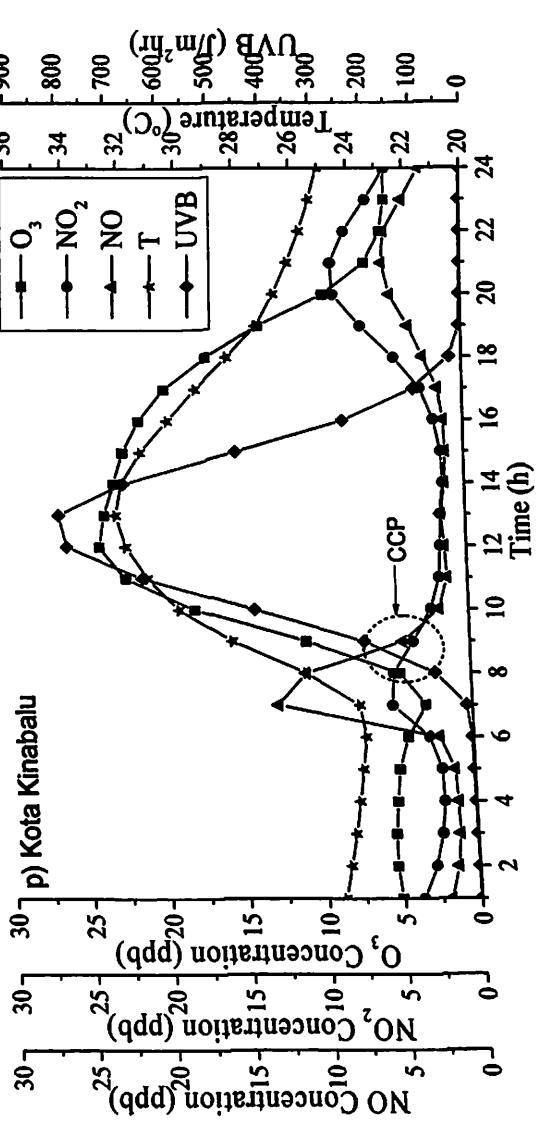


Figure 23 Composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, temperature and solar radiation in Kota Kinabalu, Jerantut and Kuching

Due to the decrease in photochemical production efficiency during late afternoon and evening, O<sub>3</sub> concentration was in a decreasing trend and at the same time NO<sub>2</sub> and NO concentrations once again increased. The increase of NO<sub>2</sub> and NO was also contributed by increase in traffic emission during evening rush hour, thus these pollutants reached their evening peaks around 9 p.m. to 10 p.m.. Ghazali et al. (2010) reported that the typical NO<sub>2</sub> diurnal trends in Malaysia showed two significant peaks early in the morning (9 a.m. to 10 a.m.) and in the evening (8 p.m. to 10 p.m.), in which the second peak is lower because of emission intensity and prevailing meteorological parameters. In the case of NO, the morning peaks are significantly higher than the evening peaks and similar trends were observed in all stations. The results are in line with the findings established by Han et al. (2011) and Banan et al. (2013) which summarized that the typical patterns of NO concentration diurnal trends are similar to that of NO<sub>2</sub> concentration, in which relatively high concentrations are observed at night and peak concentrations in the morning are attributed by vehicle emissions.

During nighttime, O<sub>3</sub> was generally low and in a stable condition because of the absence of photochemical reactions. Further reduction in nighttime O<sub>3</sub> concentrations also attributed by the deposition processes and chemical reactions (Sousa et al., 2011). The findings elucidated that nighttime O<sub>3</sub> concentrations were higher in Kemaman than in the other stations. Mean nighttime O<sub>3</sub> in Kemaman surpassed 14 ppb, whereas the constant mean concentrations ranging from 5 ppb to 6 ppb were measured in the other stations. The mean nighttime O<sub>3</sub> concentrations in Kemaman were twice as high as the concentrations in other stations, which verified the high nighttime O<sub>3</sub> occurrences in the district. Mean nighttime O<sub>3</sub> concentrations in Kemaman were relatively higher than the reported nighttime concentrations in Kolkata and Portugal with 12 ppb and 10 ppb, respectively (Ghosh et al., 2013; Kulkarni et al., 2013).

The test conducted in Kemaman revealed that NO and NO<sub>2</sub> concentrations were remarkably low with mean values of 1.1 and 4.5 ppb, respectively. The plot in Figure 4.24(d) also exhibited slightly unclear diurnal trends of NO<sub>2</sub> concentration. The maximum NO<sub>2</sub> and NO concentrations were recorded at 8 p.m. and 8 a.m. In terms of magnitude and trends of NO<sub>2</sub> and NO concentrations, similarities were

noted between Kemaman and Jerantut, which serve as background stations in Malaysia.

Nighttime  $O_3$  chemistry is predominantly controlled by NO titration. However, the mean of nighttime NO concentrations in Kemaman was 1.1 ppb, which is extremely low and contributes to the reduction of NO titration rates. Reduction of NO titrations directly affected nighttime  $O_3$  removal, thereby allowing  $O_3$  to remain in the atmosphere. Consequently, the concentration of  $NO_2$  eventually decreases with the decrease of NO titration, during which  $NO_2$  is produced. This study observed that interrupted removal reactions allowed  $O_3$  to remain in the atmosphere. Conversely, nighttime  $O_3$  concentrations in Kemaman still decreased mainly because of dry deposition and some additional chemical reactions with hydrocarbons through the reaction with  $NO_3$  (Jenkin and Clemitshaw, 2000). However, in the current studies effect of hydrocarbon is unconsidered due to limitation of monitoring data and understanding.

The shape and amplitude of  $O_3$  diurnal cycles in the study areas were strongly influenced by prevailing level of precursors and meteorological parameters. The fact that high temperature and solar radiation associated with high  $NO_2$  and NO concentration enhanced the formation of  $O_3$  to higher level cannot be denied. However, the obtained result showed that variation of the  $O_3$  concentrations in different areas are more influenced by  $NO_2$  and NO concentrations, since relatively similar UVB intensity were observed in all stations.  $O_3$  concentrations were relatively high in those stations with high  $NO_2$  and NO concentrations. Whereas, in the case of Kemaman, significantly low  $NO_2$  and NO concentrations during nighttime obstructed the  $O_3$  destruction process, thus allowed  $O_3$  to remain in the atmosphere.

#### **The $O_3$ Critical Conversion Point (CCP) from Composite Diurnal Plot**

The critical conversion points (CCP) of the  $O_3$  formations are determined using composite diurnal plot of  $O_3$ ,  $NO_2$ , NO, T and UVB in Figure 18 to Figure 23 and the time for CCP in all stations starting from 1999-2010 are shown in Table 8. CCP is

defined as the point when  $\text{NO}_2$  photolysis rate begin to surpass NO titration rate during morning which corresponds to positive ozone production rates.



Table 8 The time for critical conversion point (CCP) from 1999 to 2010 in all stations

Station	Time for Critical conversion point (CCP) (a.m.)												Average
	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	
SA	10.30	10.40	11.00	10.50	10.50	10.40	10.40	10.15	10.20	10.30	10.40	10.40	10.41
KJ	10.10	10.15	10.05	10.05	10.10	9.50	9.15	9.05	10.30	10.15	10.20	9.50	10.25
BA	9.30	9.15	9.25	9.30	9.45	9.20	9.10	9.30	9.40	9.00	8.50	9.10	9.17
KE	9.10	9.15	9.00	8.45	8.30	8.30	8.50	9.15	9.10	8.30	8.20	9.00	9.11
GB	10.20	10.30	10.45	10.40	10.50	10.50	10.45	10.35	10.40	10.55	-	-	10.41
NL	9.30	9.50	10.00	9.30	10.00	9.45	9.45	9.40	10.00	10.10	10.15	10.20	10.14
BR	9.00	9.00	9.10	9.30	9.40	9.15	9.20	9.40	9.35	9.20	9.30	9.30	9.23
IP	9.10	8.50	9.00	9.00	9.15	9.20	9.30	9.40	9.40	9.45	9.40	9.15	9.17
PR	10.20	10.00	10.05	9.40	10.05	10.00	10.15	9.20	10.05	10.00	10.30	9.45	10.30
KL	10.30	10.30	10.40	10.40	10.50	10.40	10.45	11.00	10.30	10.20	10.00	10.20	10.37
JB	10.30	10.40	10.30	10.15	10.20	10.15	10.20	10.10	10.00	10.00	10.20	10.20	10.18
TP	9.10	9.20	9.40	9.40	9.30	9.15	9.25	9.30	9.30	9.15	8.55	9.10	9.18
SJ	10.40	10.50	10.30	10.20	10.35	10.30	10.25	10.30	10.30	10.15	10.40	10.20	10.30
PG	10.15	10.30	10.20	9.55	10.00	9.50	10.00	10.00	9.55	10.00	9.50	10.00	10.30
KB	9.00	9.20	9.15	9.10	9.10	9.10	0	8.45	8.40	8.50	9.30	9.15	9.31
KK	8.00	8.30	8.00	8.00	8.10	-	0	8.00	8.10	8.10	8.20	8.10	8.09
JT	8.45	9.00	9.00	9.10	8.40	-	9.15	9.00	9.20	9.30	9.20	9.40	9.03
KC	9.55	10.00	9.40	9.45	9.20	9.10	8.40	8.15	9.45	9.40	9.20	9.50	9.23

Even though, there are crossing point between  $O_3$  and  $NO_x$  line during late afternoon, the point would not be considered as CCP as the  $O_3$  production rate are in negative rate thus will not promoting  $O_3$  accumulation. Since this study is the first to introduce the CCP, focus was more on to identify the approximate time for CCP to occur rather than the rate of CCP itself. The time for CCP is identified as the periods where high photochemical reactions rate which contributed to  $O_3$  daily accumulation. It is believed that during CCP, high photochemical reaction's efficiency was promoted by suitable amount of precursor and sunlight radiation.

The result demonstrated that the CCP were measured approximately in between 8 a.m. to 11 a.m. in all study locations with an average time around 10 a.m.. The earliest CCP was measured in Kota Kinabalu at around 8.00 a.m. to 8.30 a.m. and followed by Kota Bharu and Kemaman with all recorded CCP around 9 a.m. Meanwhile, the latest CCP was observed in stations such as Shah Alam, Klang, Johor Bahru and Gombak in between 10.20 a.m. to 11 a.m..

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. 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 CCP is reached,  $O_3$  will show increment trends, while  $NO_2$  and  $NO$  will showed decrement trends. It was observed that,  $O_3$  concentrations in stations where their CCP occur around 10 a.m. and later are relatively higher than the stations that have their CCP earlier. The mean of  $O_3$  concentrations for Shah Alam, Klang, Johor Bahru and Gombak was around 20 ppb, while for Kota Kinabalu, Kota Bharu and Jerantut was around 10 ppb. This finding opened the possibilities to use time for CCP to give some indications regarding  $O_3$  concentration levels at one particular location.

$NO$  concentration play an important role in CCP determination. High concentrations of  $NO$  are released by the anthropogenic sources such as motor vehicles and industrial establishments in the morning that causes  $NO$  to reach their peak

concentration around 8 a.m. to 9 a.m.. Due to the high concentration of NO, the NO titration rate will also increase, hence increasing O<sub>3</sub> scavenge which will profoundly reduce the O<sub>3</sub> concentrations. The minimum O<sub>3</sub> concentrations in Shah Alam, Klang, Gombak and Seberang Jaya was 3.03 ppb, 4.31 ppb, 1.82 ppb and 1.98 ppb, respectively. However, high NO titration will ultimately produce high concentration of NO<sub>2</sub>, which is later converted into O<sub>3</sub>. In these stations, the time for CCP will be later because NO<sub>2</sub> photolysis rate will take longer to surpass NO titration.

Alternatively, the time for CCP will be faster when the NO titration rate is lower. According to the results, there is only a 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 barely affected the CCP variations. In addition, the usage of long period of data (12 years) caused the effect of weather condition to be insignificant.

### **Next Hour Ozone Prediction Model**

Regression analysis is utilized to examine the relationship between O<sub>3</sub> and several independent variables such as concentration of NO<sub>2</sub>, NO, SO<sub>2</sub>, CO, temperature, relative humidity, wind speed and UVB. Regression analysis was also used to develop next hour prediction models based on these independent variables to predict next hour O<sub>3</sub> concentrations. In this study, current O<sub>3</sub> concentrations were added in the models as an extra independent variables considered as the lagged concentrations. According to Comrie (1997), O<sub>3</sub> concentrations are partially dependent on the previous O<sub>3</sub> concentrations and showed serial correlation. In contrast, regression model assume the observations as statistically independent events. To avoid this problem and still incorporating the importance of persistence, Comrie (1997) suggested to use a lagged O<sub>3</sub> concentration as an additional predictor variable in the model especially for prediction purpose. Barrero et al. (2006) also used the same strategy in order to improve the accuracy of prediction.

### Group of Monitoring Stations Based on Ozone Concentrations

In this study, O<sub>3</sub> prediction models are developed on several group of monitoring stations that were based on DoE classification, ranking of means and cluster analysis groups. Industrial, urban, sub-urban and background are DoE based group while ranking of means and cluster groups are developed based on O<sub>3</sub> concentrations analysis.

The summary of the group of monitoring stations in regards of DoE classifications and newly developed using ranking of means and cluster analysis are depicted in Table 9. For groups based on the DoE classifications, the same groups of monitoring stations were used for all ranges of time. Meanwhile, for ranking of means and cluster groups, there was variability in the selection of monitoring stations in each group. The summary table illustrated that six groups from ranking and cluster shared the same monitoring stations. The group are G3-D/C2-D, G1-N/C2-N and G2-N/C2-N. Further analysis in the next subsections were performed based on these groups.

Table 9 Summary of the monitoring stations group based on DoE, ranking of means and cluster analysis during different ranges of time

DoE				Rank of means				Cluster analysis			
AT	DT	NT	CT	AT	DT	NT	CT	AT	DT	NT	CT
<u>IN</u>	<u>IN</u>	<u>IN</u>	<u>IN</u>	<u>G1</u>	<u>G1</u>	<u>G1</u>	<u>G1</u>	<u>C1</u>	<u>C1</u>	<u>C1</u>	<u>C1</u>
PG	PG	PG	PG	KC	KC	GB	SJ	PG	KC	KC	SJ
KE	KE	KE	KE	KK	JT	KK	KC	JB	KK	KK	TP
PR	PR	PR	PR	JT	KB	TP	JT	JT	KB	JT	PR
KC	KC	KC	KC	KB	KK	JT	GB	KB	JT	JB	JT
BR	BR	BR	BR	PG	PG	SJ	IP	KC	PG	TP	IP
IP	IP	IP	IP	SJ		JB	TP	KK	TP	GB	KC
NL	NL	NL	NL	JB		KC		IP	SJ	IP	PG
TP	TP	TP	TP	TP	<u>G2</u>	KJ	<u>G2</u>	KL	KE	PG	JB

<u>UB</u>	<u>UB</u>	<u>UB</u>	<u>UB</u>	PR	SJ	PG	PR	PR	BR	KB	BR
KB	KB	KB	KB	KL	TP	SA	PG	SJ	JB	KL	NL
KJ	KJ	KJ	KJ	IP	JB	IP	KB	TP	SP	SJ	BA
SA	SA	SA	SA	BR	SP	KL	NL	BA	KL	KJ	KB
KK	KK	KK	KK	GB	BR	KB	JB	BR	IP	SA	KK
GB	GB	GB	GB	NL	KL	NL	KL	NL	BA	BR	KE
KL	KL	KL	KL		IP	SP	BR	KE	NL	NL	
JB	JB	JB	JB	<u>G2</u>	KE	BR	KJ		GB	SP	<u>C2</u>
				KE	NL		SA	<u>C2</u>			GB
				KJ	GB	<u>G2</u>	KK	GB	<u>C2</u>	<u>C2</u>	KL
<u>SU</u>	<u>SU</u>	<u>SU</u>	<u>SU</u>	BA	BA	BA	BA	KJ	KJ	KE	KJ
SJ	SJ	SJ	SJ	SA		KE	KE	SA	SA	BA	SA
BA	BA	BA	BA		<u>G3</u>						
<u>BG</u>	<u>BG</u>	<u>BG</u>	<u>BG</u>		<u>D</u>						
JT	JT	JT	JT		KJ						
					SA						

Note: IN is industrial; UB is urban; SU is sub urban; BG is background; G is group based on ranking of mean; C is group based on cluster analysis when; AT is 'daily'; DT is daytime; NT is nighttime; CT is critical conversion time

### Variables Reduction using PCA

PCA was applied on ten variables of each group to determine the major contributors to O<sub>3</sub> variations in each group, as well as to reduce the number of independent variables. According to Özbay et al. (2011) reduction in the number of independent variables will avoid the multicollinearity problems in a prediction model. Similar independent variables to MLR analysis were used in order to compare the efficiency of MLR and PCR models. Since sunlight was absent during NT, UVB was excluded during NT analysis. Table 10 summarized the PCA results during difference time range and group of monitoring stations.

Table 10 Principal Components Analysis after varimax rotation for respective group of monitoring stations during AT, DT, NT and CT

Group	KMO	B	ZV (%)	EV (%)	Variables	ZVal (%)
<b>Daily (AT)</b>						
IN-AT	0.853	0.001	74.22	PC1	CO NO <sub>3</sub> RH T UVB WS	73.92
				PC2	NO <sub>2</sub> SO <sub>2</sub>	
UB-AT	0.852	0.001	77.10	PC1	O <sub>3</sub> RH T UVB WS	77.14
				PC2	CO NO NO <sub>2</sub> SO <sub>2</sub>	
SU-AT	0.847	0.001	71.71	PC1	O <sub>3</sub> RH T UVB WS	71.38
				PC2	CO NO NO <sub>2</sub> SO <sub>2</sub>	
BG-AT	0.776	0.001	70.26	PC1	NO O <sub>3</sub> RH T UVB WS	71.22
				PC2	CO NO	
				PC3	SO <sub>2</sub>	
G1-AT	0.849	0.001	78.26	PC1	O <sub>3</sub> RH T UVB WS	78.12
				PC2	CO NO NO <sub>2</sub> SO <sub>2</sub>	
G2-AT	0.877	0.001	74.09	PC1	O <sub>3</sub> RH T UVB WS	74.12
				PC2	CO NO NO <sub>2</sub> SO <sub>2</sub>	
C1-AT	0.854	0.001	77.87	PC1	O <sub>3</sub> RH T UVB WS	77.74
				PC2	CO NO NO <sub>2</sub> SO <sub>2</sub>	
C2-AT	0.831	0.001	74.07	PC1	NO O <sub>3</sub> RH T UVB WS	77.18
				PC2	CO NO <sub>2</sub> SO <sub>2</sub>	

Daytime (DT)							
IN-DT	0.869	0.001	72.21	PC1	58.97	CO NO <sub>2</sub> O <sub>3</sub> RH T UVB	72.65
						WS	
				PC2	13.23	NO <sub>2</sub> SO <sub>2</sub>	
UB-DT	0.850	0.001	73.44	PC1	58.44	CO NO <sub>2</sub> O <sub>3</sub> RH T UVB	73.98
						WS	
				PC2	15.24	NO <sub>2</sub> SO <sub>2</sub>	
SU-DT	0.861	0.001	71.13	PC1	58.27	NO <sub>2</sub> O <sub>3</sub> RH T UVB	71.28
				PC2	12.86	CO NO <sub>2</sub> SO <sub>2</sub> WS	
BG-DT	0.784	0.001	71.21	PC1	43.62	NO <sub>2</sub> O <sub>3</sub> RH T UVB WS	70.21
				PC2	16.72	CO NO <sub>2</sub>	
				PC3	10.87	SO <sub>2</sub>	
G1-DT	0.857	0.001	71.62	PC1	56.84	CO NO <sub>2</sub> O <sub>3</sub> RH T UVB	70.04
						WS	
				PC2	14.78	NO <sub>2</sub> SO <sub>2</sub>	
G2-DT	0.873	0.001	74.69	PC1	61.48	CO NO <sub>2</sub> O <sub>3</sub> RH T UVB	75.09
						WS	
				PC2	13.20	NO <sub>2</sub> SO <sub>2</sub>	
G3-DT	0.823	0.001	69.38	PC1	52.29	O <sub>3</sub> RH T UVB WS	69.63
				PC2	17.09	CO NO NO <sub>2</sub> SO <sub>2</sub>	
C1-DT	0.873	0.001	77.50	PC1	64.10	CO NO <sub>2</sub> O <sub>3</sub> RH T UVB	76.60
						WS	
				PC2	13.40	NO <sub>2</sub> SO <sub>2</sub>	
C2-DT	0.796	0.001	65.88	PC1	48.19	CO O <sub>3</sub> RH T UVB WS	69.63
				PC2	17.69	NO NO <sub>2</sub> SO <sub>2</sub>	

Nighttime (NT)							
IN-NT	0.733	0.001	64.29	PC1	38.19	CO NO NO <sub>2</sub> SO <sub>2</sub> T	64.75
				PC2	26.10	O <sub>3</sub> RH WS	
UB-NT	0.708	0.001	63.73	PC1	35.27	CO NO NO <sub>2</sub> SO <sub>2</sub> T	64.26
				PC2	28.46	O <sub>3</sub> RH WS	
SU-NT	0.712	0.001	63.36	PC1	34.18	CO NO NO <sub>2</sub> SO <sub>2</sub>	64.51
				PC2	29.18	O <sub>3</sub> RH T WS	
BG-NT	0.644	0.001	61.24	PC1	30.31	NO O <sub>3</sub> WS	60.88
				PC2	17.41	CO NO <sub>2</sub>	
				PC3	13.53	RH SO <sub>2</sub> T	
G1-NT	0.740	0.001	67.22	PC1	40.52	CO NO NO <sub>2</sub> SO <sub>2</sub>	67.56
				PC2	26.70	O <sub>3</sub> RH T WS	
G2-NT	0.656	0.001	67.97	PC1	30.98	O <sub>3</sub> RH WS	66.88
				PC2	24.83	CO NO SO <sub>2</sub>	
				PC3	12.16	NO <sub>2</sub> T	
C1-NT	0.740	0.001	67.22	PC1	40.52	CO NO NO <sub>2</sub> SO <sub>2</sub> T	67.56
				PC2	26.70	O <sub>3</sub> RH WS	
C2-NT	0.656	0.001	67.97	PC1	30.98	O <sub>3</sub> RH WS	66.88
				PC2	24.83	CO NO SO <sub>2</sub>	
				PC3	12.16	NO <sub>2</sub> T	



Note: B is Bartlett's test of sphericity;  $\Sigma V$  is total variance explained; EV is variance for each PC;  $\Sigma Val$  is total variance for validation

Critical Conversion Time (CT)			PC1	PC2	PC3	$\Sigma V$	B	EV
IN-CT	CO NO <sub>3</sub> T UVB WS	44.55	18.16	NO <sub>2</sub> SO <sub>2</sub>	14.54	77.25	0.001	0.775
UB-CT	RH	14.54	PC3	RH	76.31	0.001	0.744	0.001
	CO NO T UVB WS	42.14	PC2	NO <sub>2</sub> O <sub>3</sub>				
	CO NO T UVB WS	42.14	PC1	CO NO T UVB WS				
SU-CT	CO NO T UVB WS	35.26	PC1	CO NO T UVB WS	66.84	0.001	0.643	0.001
	RH SO <sub>2</sub>	16.68	PC2	RH SO <sub>2</sub>				
	NO <sub>2</sub> O <sub>3</sub>	14.90	PC3	NO <sub>2</sub> O <sub>3</sub>				
BG-CT	NO RH T UVB WS	39.08	PC1	NO RH T UVB WS	68.34	0.001	0.666	0.001
	CO NO <sub>2</sub> O <sub>3</sub>	18.11	PC2	CO NO <sub>2</sub> O <sub>3</sub>				
	SO <sub>2</sub>	11.24	PC3	SO <sub>2</sub>				
GI-CT	CO NO T UVB WS	35.74	PC1	CO NO T UVB WS	79.39	0.001	0.688	0.001
	NO <sub>2</sub>	19.20	PC2	NO <sub>2</sub>				
	RH SO <sub>2</sub>	17.48	PC3	RH SO <sub>2</sub>				
	O <sub>3</sub>	8.28	PC4	O <sub>3</sub>				
G2-CT	CO NO O <sub>3</sub> T UVB WS	52.59	PC1	CO NO O <sub>3</sub> T UVB WS	83.64	0.001	0.804	0.001
	RH SO <sub>2</sub>	13.31	PC2	RH SO <sub>2</sub>				
	NO <sub>2</sub>	12.74	PC3	NO <sub>2</sub>				
C1-CT	CO NO RH T UVB WS	54.66	PC1	CO NO RH T UVB WS	82.84	0.001	0.816	0.001
	WS		PC2	NO <sub>2</sub> SO <sub>2</sub>				
	O <sub>3</sub>	12.35	PC3	O <sub>3</sub>				
C2-CT	CO NO O <sub>3</sub> RH T UVB WS	56.24	PC1	CO NO O <sub>3</sub> RH T UVB WS	78.34	0.001	0.867	0.001
	NO <sub>2</sub> SO <sub>2</sub>	22.11	PC2	NO <sub>2</sub> SO <sub>2</sub>				
		81.69						
		83.20						
		84.6						
		80.14						
		68.22						
		63.08						
		74.15						
		77.05						

The sufficiency of the monitoring records for factor analysis was assessed using Kaiser-Meyer-Olkin (KMO) test and Bartlett's test of sphericity. The results indicated that the KMO value ranged from 0.643 (SU-CT) to 0.877 (G2-AT), which were higher than 0.5, showed that the data was adequate for the PCA. Meanwhile, *p*-value for Bartlett's test of sphericity was 0.001 at all stations which was lower than 0.05, indicated that the high degree of relationship between the selected variables and the dataset are suitable for PCA (Ul-Saufie et al., 2013).

The results showed that the total variance explained during AT, DT, NT and CT ranged from 70.26 – 78.26%, 69.63 – 76.6%, 60.88 – 67.56% and 63.08 – 84.6%, respectively. The highest total variance was 84.6% which was measured during CT for G2-CT group, while the lowest percentage was 60.88% which was measured during NT for BG-NT.

The percentage of total variance indicated that the ability of the selected variables in explaining variations in O<sub>3</sub> concentration (Abdul-Wahab et al., 2005; Özbay et al., 2011) which is the best using CT followed by AT, DT and NT with average of total variance explained was 76.52%, 75.10%, 72.12% and 65.14%, respectively. Higher percentage of total variance was measured for ranking of mean and cluster group compared to the DoE group which were observed during all time periods. The results were due the consistency in O<sub>3</sub> concentration variations in both ranking of mean and cluster groups, since both groups were developed based on O<sub>3</sub> concentrations.

In an extent to variable reduction capabilities, PCA was also capable to determine the contribution of the selected variables towards the variation in O<sub>3</sub> concentrations. PCA divided the variables mostly into two PCs (22 groups), three PCs (10 groups) and four PCs for only one group which was G1-CT based on eigenvalue larger than 1. In PCA, it is known that PC1 contributed the highest percentage of variance followed by the other PCs (Dominick et al., 2012) and the significant variables for each PC is determined based on factor loading value. In this study, only factor

loadings that are higher than 0.4 was considered, while the factor loading lower than 0.4 was removed from the analysis.

### **Multiple Linear Regression Models**

In ambient air, the inter-relationship between O<sub>3</sub> and the contributing factors are very complex, however, MLR was proven to be successfully used to predict as well as to evaluate the dependency of the O<sub>3</sub> concentration towards the primary pollutants and meteorological parameters (Abdul Wahab et al., 2005; Barerro et al., 2006; Ghazali et al., 2010). Summary of stepwise MLR to predict next hour O<sub>3</sub> concentrations for all groups during different time range is shown in Table 11. Stepwise MLR was chosen in this study in order to produce parsimonious model that is able to maximize prediction accuracy with minimal number of predictor variables. Abdul-Wahab et al. (2005) and Ghazali et al. (2010) also have performed similar procedures to select the most significant variables in explaining O<sub>3</sub> variations during daytime and nighttime.

Result exhibited that the selected variables did not encounter any multicollinearity problems indicated by VIF values which was less than 10. During the model development stage, the variables showed VIF values larger than 10 was removed from the models to avoid multicollinearity problem. Based on Durbin-Watson (*d*) values, the developed models also did not encounter any autocorrelations problems as the VIF values were significantly higher than the upper limit (*d<sub>u</sub>*) of the critical value for Durbin-Watson test which was 1.87. For all of the developed models, the *d* values was ranged in between 1.938 (BG-N) to 2.027 (G1-A).

The performance of the developed MLR models were investigated using adjusted coefficient of determination ( $\bar{R}^2$ ) value to express the fit of the models. Abdul-Wahab et al. (2005) pointed that  $\bar{R}^2$  is more accurate than  $R^2$  as it considered the number of independent variables. Adjusted  $R^2$  is a modified version of  $R^2$  that compares the explanatory power of regression models that contain different number of predictors. The adjusted  $R^2$  increase only if the addition of new variable capable to

improve the model performance. Hence in this study,  $\bar{R}^2$  was used to indicate the performance of the regression models.

Table 11 Summary of MLR models for O<sub>3</sub> concentration prediction for all groups during different time range

Group	$\bar{R}^2$	Models	<i>d</i>	VIF
<b>'Daily' (AT)</b>				
IN-AT	0.922	$O_{3,t} = 18.239 + 0.718O_3 + 0.825WS - 0.206NO$ $+ 0.164NO_2 - 0.215RH - 0.006UVB -$ $0.130SO_2$	1.998	1.152 – 6.193
UB-AT	0.919	$O_{3,t} = -28.374 + 0.703O_3 + 1.173WS - 0.114NO$ $- 0.013UVB + 1.200T - 0.069NO_2$	2.007	1.479 – 8.551
SU-AT	0.864	$O_{3,t} = 15.633 + 0.750O_3 + 0.888WS - 0.098NO$ $- 0.184RH - 0.007UVB$	2.000	1.407 – 3.570
BG-AT	0.882	$O_{3,t} = 29.160 + 0.661O_3 - 0.336RH -$ $0.009UVB - 0.608NO + 0.005CO + 0.515WS$ $+ 0.413SO_2$	2.027	1.039 – 2.469
G1-AT	0.934	$O_{3,t} = 8.810 + 0.738O_3 + 1.207WS - 0.176NO -$ $0.007UVB - 0.123RH + 0.136NO_2$	2.007	1.442 – 7.027
G2-AT	0.890	$O_{3,t} = 27.418 + 0.722O_3 + 1.077WS - 0.183NO$ $- 0.282RH - 0.011UVB - 0.526SO_2$	2.004	1.121 – 5.548
C1-AT	0.935	$O_{3,t} = 9.498 + 0.726O_3 + 1.200WS - 0.205NO$ $- 0.006UVB - 0.128RH + 0.154NO_2 -$ $0.096SO_2$	2.005	1.194 – 6.891
C2-AT	0.888	$O_{3,t} = 26.714 + 0.800O_3 + 1.423WS - 0.075NO$ $- 0.018UVB - 0.286RH - 0.461SO_2 -$	2.001	1.397 –

		0.028NO <sub>2</sub>		5.002
<b>Daytime (DT)</b>				
IN-DT	0.857	$O_{3,t} = 3.635 - 0.344RH + 0.388O_3 - 0.405NO - 0.007UVB + 0.990T + 0.847WS + 0.007CO + 0.333NO_2 - 0.198SO_2$	1.984	1.135 - 8.044
UB-DT	0.839	$O_{3,t} = -43.797 + 1.889T + 0.557O_3 - 0.203NO - 0.015UVB + 1.286WS - 0.060NO_2$	1.987	1.466 - 5.356
SU-DT	0.786	$O_{3,t} = 31.803 + 0.588O_3 + 1.057WS - 0.341RH - 0.214NO - 0.10UVB - 0.095NO_2 + 0.136SO_2 + 0.001CO$	2.013	1.163 - 4.409
BG-DT	0.849	$O_{3,t} = 34.807 + 0.549O_3 - 0.410RH - 0.010UVB + 0.788WS + 0.006CO - 0.704NO + 0.348NO_2 + 0.623SO_2$	2.002	1.037 - 2.596
G1-DT	0.831	$O_{3,t} = 5.532 - 0.244RH + 0.469O_3 - 0.309NO - 0.005UVN + 0.604WS + 0.281NO_2 + 0.614T + 0.003CO$	1.997	1.533 - 7.581
G2-DT	0.856	$O_{3,t} = -25.231 + 1.398T + 1.543WS + 0.429O_3 - 0.009UVB - 0.348NO + 0.006CO - 0.134RH - 0.146SO_2 + 0.107NO_2$	1.978	1.176 - 9.498
<b>'Daily' (AT)</b>				
G3-DT	0.802	$O_{3,t} = -49.706 + 0.638O_3 + 2.253T - 0.268NO - 0.021UVB + 1.122WS - 0.119NO_2 - 0.060SO_2$	2.004	1.358 - 3.502
C1-DT	0.875	$O_{3,t} = -31.382 + 1.693T + 1.201WS + 0.387O_3 - 0.010UVB - 0.363NO + 0.008CO - 0.135RH - 0.143SO_2$	1.989	1.104 - 8.795
C2-DT	0.802	$O_{3,t} = -49.706 + 0.638O_3 + 2.253T - 0.268NO - 0.021UVB + 1.122WS - 0.119NO_2 -$	2.004	1.358 -

		0.060SO <sub>2</sub>		3.502
<b>Nighttime (NT)</b>				
IN-NT	0.737	$O_{3,t} = 7.154 + 0.391O_3 - 0.262RH + 1.130WS + 0.676T - 0.156NO - 0.247SO_2 + 0.001CO + 0.052NO_2$	1.982	1.246 – 2.983
UB-NT	0.662	$O_{3,t} = -0.190 + 1.140WS + 0.395O_3 + 0.698T - 0.093NO - 0.178RH - 0.205SO_2 + 0.043NO_2$	2.013	1.206 – 2.625
SU-NT	0.620	$O_{3,t} = 3.242 + 0.471O_3 - 0.237RH - 0.115NO + 0.821T + 0.629WS - 0.228SO_2$	2.009	1.200 – 2.399
BG-NT	0.684	$O_{3,t} = 27.712 + 1.808WS - 0.300RH + 0.275O_3 - 0.600NO + 0.002CO + 0.128NO_2$	1.938	1.096 – 1.592
G1-NT	0.758	$O_{3,t} = 7.696 + 1.215WS + 0.331O_3 - 0.240RH - 0.167NO + 0.591T - 0.313SO_2 + 0.070NO_2 + 0.001CO$	1.985	1.320 – 3.865
G2-NT	0.701	$O_{3,t} = 18.516 - 0.432RH + 0.365O_3 + 1.318WS + 0.876T - 0.367NO + 0.002CO$	1.961	1.357 – 2.342
C1-NT	0.758	$O_{3,t} = 7.696 + 1.215WS + 0.331O_3 - 0.240RH - 0.167NO + 0.591T - 0.313SO_2 + 0.070NO_2 + 0.001CO$	1.985	1.320 – 3.865
C2-NT	0.701	$O_{3,t} = 18.516 - 0.432RH + 0.365O_3 + 1.318WS + 0.876T - 0.367NO + 0.002CO$	1.961	1.357 – 2.342
<b>Critical Conversion Time (CT)</b>				
IN-CT	0.759	$O_{3,t} = -6.609 + 0.016UVB + 1.008WS - 0.236NO + 0.408T + 0.233NO_2 - 0.109SO_2 + 0.005RH$	2.024	1.311 – 2.423

UB-CT	0.832	$O_{3,t} = -35.663 + 1.642T + 0.016UVB - 0.144NO - 0.087O_3 + 0.011RH + 0.001CO + 0.301WS + 0.052NO_2$	2.003	1.101 - 6.291
SU-CT	0.593	$O_{3,t} = -7.391 + 0.023UVB + 0.091O_3 - 0.100NO + 0.534T + 0.209NO_2 - 0.002CO + 0.288WS$	2.001	1.262 - 3.446
BG-CT	0.724	$O_{3,t} = 50.599 - 0.560RH + 1.012WS + 0.505NO_2 - 0.480NO + 0.006CO + 1.003SO_2$	1.994	1.036 - 1.966
G1-CT	0.785	$O_{3,t} = -8.627 + 0.021UVB + 0.379T - 0.098NO + 0.269NO_2 + 0.678WS - 0.208SO_2 - 0.031O_3 + 0.001CO$	1.987	1.154 - 4.775
G2-CT	0.841	$O_{3,t} = -28.739 + 1.269T + 1.053WS - 0.206NO + 0.016UVB - 0.152SO_2 + 0.003CO + 0.006RH$	1.997	1.553 - 5.341
C1-CT	0.848	$O_{3,t} = -0.036 - 0.188RH - 0.219NO + 0.662T + 1.147WS + 0.004CO + 0.012UVB - 0.228SO_2 + 0.204NO_2$	1.981	1.284 - 8.084
C2-CT	0.808	$O_{3,t} = -1.586 + 1.342T + 0.016UVB - 0.120NO - 0.108O_3 - 0.253RH - 0.266WS - 0.045NO_2 + 0.001CO$	1.995	1.378 - 9.079

The  $\bar{R}^2$  values for AT, DT, NT and CT was in range of 0.882 – 0.935, 0.786 – 0.875, 0.620 – 0.758, 0.593 – 0.848, respectively. Results showed that MLR models using AT data produced the highest accuracy with 0.935 (G1-AT-MLR) which was closest to the value of one. It gave an indication that the selected variables were able to explain variation in  $O_3$  concentrations during AT up to 93.5%. The average  $\bar{R}^2$  value for all developed models during AT was 0.904%. Conversely, the values of adjusted coefficient of determination ( $\bar{R}^2$ ) during DT were slightly lower than during AT with an average of  $\bar{R}^2$  of 0.833%. For all groups except for SU-DT-MLR, the  $\bar{R}^2$  value is

surpassed 0.8. The maximum  $\bar{R}^2$  during DT was 0.875 that was measured for C1-DT-MLR, while the minimum was 0.796 (SU-DT-MLR).

The efficiency of MLR models during NT were significantly lower as compared to the other time period. During NT, value of  $\bar{R}^2$  was ranged from 0.620 (SU-NT-MLR) to 0.758 (G1/CI-N-MLR). The same models were used for G1-NT-MLR/C1-NT-MLR and G2-NT-MLR/C2-NT-MLR, since the monitoring stations grouped in the groups that are similar. Significantly lower in  $\bar{R}^2$  value during NT was perhaps contributed by the suppression of UVB data from NT models. Comparatively,  $\bar{R}^2$  during the CT was very similar to DT, even there were only four hours of monitoring data used in representing the CT, compared to 12 hours used for DT. The  $\bar{R}^2$  value during CT was in the range of 0.593 (SU-CT-MLR) to 0.848 (UB-CT-MLR).

It was observed that  $\bar{R}^2$  was relatively higher for MLR models using ranking of means and cluster group compared to the original group by DoE. The highest  $\bar{R}^2$  value for all time period was from either ranking of means or from the cluster group. This finding proved than groups that were developed using the  $O_3$  concentrations were more suitable in explaining variations in  $O_3$  concentrations compared to groups that developed based on the locations of the monitoring stations.

The results illustrated that, the current  $O_3$  concentration was the main factor in predicting next hour  $O_3$  variations during AT for all groups. Ghazali et al. (2010) also reported similar findings which were relevant since hourly  $O_3$  concentration variations was quite small especially during NT. WS was also another important variable in MLR-AT, since the increased WS will directly lower  $O_3$  resulted by the dispersal effect (Ul-Saufie et al., 2013). Elminir (2005) also selected wind as an important meteorological parameters that influence the behaviour of air pollutants. Barrero et al. (2006) added that WS showed high correlations with all type of air pollutants which was probably due to the direct dilution effects.



MLR-AT also listed NO, NO<sub>2</sub> and UVB which were the main ingredients of O<sub>3</sub> photochemical reactions as important factors in this time period models. However, for all MLR models during AT, T was excluded from most of the models due to VIF values above 10 which indicated multicollinearity problem. This might be due a high correlation between T and UVB.

It was observed that, during DT, RH, current O<sub>3</sub>, WS, UVB, NO, NO<sub>2</sub> and CO played significant roles in explaining the variations of O<sub>3</sub> concentrations since these variables existed in most of the DT models. DT models such as IN-DT-MLR and G1-DT-MLR ranked RH as their most important factor, while models such as UB-DT-MLR, G2-DT-MLR, and C1-DT-MLR placed T as their important factor. High temperature provided an indication of conducive environmental condition for O<sub>3</sub> formation and accumulation. In addition, O<sub>3</sub> showed a high positive correlation with temperature especially during DT. Meanwhile, O<sub>3</sub> showed inverse relationship with RH. For the rest of the MLR-DT models, the most important factor was current O<sub>3</sub>. Even though variables such as NO, NO<sub>2</sub> and UVB was directly linked to O<sub>3</sub> photochemical reactions, other variables such as WS, RH and CO also played significant roles in daytime O<sub>3</sub> chemistry.

It was established that the O<sub>3</sub> photochemical reactions only occurred during DT. During NT, O<sub>3</sub> was removed from the atmosphere through several pathways such as chemical reaction, depositions and transportations. Results suggested that NT O<sub>3</sub> was highly influenced by the current O<sub>3</sub>, WS, RH as well as NO concentrations. The current O<sub>3</sub> represents the retained O<sub>3</sub> from DT, while WS is a significant air pollutant dispersion and transport agent (Camalier et al., 2007). RH represents the humidity in the air and high humidity is associated with rain which capable in promoting wet deposition, thus reducing O<sub>3</sub> concentrations (Toh et al., 2013). Reactions between O<sub>3</sub> and NO is the most significant path where O<sub>3</sub> is removed from the atmosphere and during NT, NO<sub>2</sub> also acts as O<sub>3</sub> destruction agents which converts O<sub>3</sub> to HNO<sub>3</sub> (Jenkin and Clemitshaw, 2000).

O<sub>3</sub> exhibited unique trends during CT (8 a.m. – 11 a.m.) as compared to other primary air pollutants. During this period, O<sub>3</sub> reached its minimum point which was usually measured at 8 a.m. and after that O<sub>3</sub> started to increase and accumulate to a higher concentration due to high rate of photochemical reactions. During this period, MLR identified UVB, WS, NO, NO<sub>2</sub> and the current O<sub>3</sub> as the most significant factor in predicting O<sub>3</sub> concentrations. In the morning, NO concentration was relatively high due to vehicle emissions during rush hour, thus significantly promoted NO nitration which produced NO<sub>2</sub> concentrations. An increase in NO<sub>2</sub> will directly enhance NO<sub>2</sub> photolysis and O<sub>3</sub> production with the availability of UVB. Higher NO<sub>2</sub> photolysis rate than NO titration rate will eventually promote O<sub>3</sub> accumulations and according to Ghazali et al. (2010) this situation is enhanced when WS is relatively low (less than 3 m/s).

### Principal Component Regression Models

The capabilities of PCA to transform the original variables into new, uncorrelated variables (PCs) were utilized to form new linear combinations of the selected variables. The new developed PCs were then used as the input to MLR to form the PCR, which is less complex than the MLR models due to less number of input variables (Kumar and Goyal, 2011). Nevertheless, all assumptions for OLS still need to be fulfilled. The summary of PCR models at different time range is shown in Table 12.

Table 12 Summary of PCR models for O<sub>3</sub> concentration prediction for all groups during different time range

Group	$\bar{R}^2$	Models	$d$	Range of VIF
<b>'Daily' (AT)</b>				
IN-AT	0.590	$O_{3,t} = 15.896 + 0.036PC1 - 0.008PC2$	2.007	1.310
UB-AT	0.679	$O_{3,t} = 16.341 + 0.049PC1 - 0.009PC2$	2.011	1.315

SU-AT	0.488	$O_{3,t} = 20.716 + 0.037PC1 - 0.011PC2$	1.993	1.265
BG-AT	0.417	$O_{3,t} = 1.131 + 0.039PC1 + 0.023PC2 + 2.043PC3$	2.027	1.006 - 1.047
G1-AT	0.666	$O_{3,t} = 14.061 + 0.039PC1 - 0.007PC2$	2.013	1.365
G2-AT	0.625	$O_{3,t} = 23.215 + 0.052PC1 - 0.014PC2$	2.000	1.230
C1-AT	0.651	$O_{3,t} = 15.157 + 0.037PC1 - 0.007PC2$	2.011	1.362
C2-AT	0.631	$O_{3,t} = 19.016 + 0.067PC1 - 0.008PC2$	2.006	1.249
<b>Daytime (DT)</b>				
IN-DT	0.487	$O_{3,t} = 24.110 + 0.024PC1 + 0.328$	2.019	1.155
UB-DT	0.340	$O_{3,t} = 42.570 + 0.0004PC1 - 0.045PC2$	2.000	1.341
SU-DT	0.454	$O_{3,t} = 28.227 + 0.027PC1 - 0.021PC2$	2.007	1.522
BG-DT	0.347	$O_{3,t} = 1.228 + 0.035PC1 + 0.025PC2 - 0.459PC3$	2.000	1.018 - 1.046
G1-DT	0.440	$O_{3,t} = 19.424 + 0.017PC1 + 0.167PC2$	2.004	1.166
G2-DT	0.556	$O_{3,t} = 25.655 + 0.027PC1 + 0.486PC2$	2.007	1.302
G3-DT	0.462	$O_{3,t} = 27.888 + 0.059PC1 - 0.014PC2$	1.997	1.191
C1-DT	0.564	$O_{3,t} = 24.403 + 0.025PC1 + 0.461PC2$	2.005	1.315
C2-DT	0.356	$O_{3,t} = 27.841 - 0.061PC1 - 0.018PC2$	2.003	1.129
<b>Nighttime (NT)</b>				
IN-NT	0.546	$O_{3,t} = 4.498 - 0.638PC2 + 0.014PC1$	1.993	1.142
UB-NT	0.519	$O_{3,t} = 27.969 + 0.392PC2 + 0.003PC1$	1.997	1.328
SU-NT	0.555	$O_{3,t} = 33.144 - 0.518PC2 + 0.05PC1$	2.002	1.229
BG-NT	0.612	$O_{3,t} = 12.921 + 0.633PC1 + 0.402PC3 + 0.002PC2$	1.961	1.021 - 1.177
G1-NT	0.654	$O_{3,t} = 45.258 + 0.657PC2 - 0.002PC1$	1.976	1.020
G2-NT	0.657	$O_{3,t} = 44.433 + 0.694PC1 + 0.694PC3 - 0.002PC2$	1.985	1.022 - 1.175
C1-NT	0.654	$O_{3,t} = 45.258 + 0.657PC2 - 0.002PC1$	1.976	1.020
C2-NT	0.657	$O_{3,t} = 44.433 + 0.694PC1 + 0.694PC3 - 0.002PC2$	1.985	1.022 - 1.175

Critical Conversion Time (CT)				
IN-CT	0.642	$O_{3,t} = 6.704 + 0.034PC1 - 0.006PC2 - 0.020PC3$	2.024	1.028 - 1.655
UB-CT	0.571	$O_{3,t} = 18.544 + 0.015PC1 + 0.119PC2 + 0.008PC3$	2.018	1.003 - 1.029
SU-CT	0.414	$O_{3,t} = 15.845 - 0.011PC1 + 0.214PC3 + 0.017PC2$	2.014	1.033 - 1.058
BG-CT	0.694	$O_{3,t} = 55.928 + 1.012PC3 + 0.007PC2 + 0.014PC1$	2.051	1.156 - 4.924
G1-CT	0.613	$O_{3,t} = 8.159 + 0.015PC1 + 0.792PC2 - 0.047PC4 + 0.006PC3$	2.011	1.036 - 1.109
G2-CT	0.783	$O_{3,t} = 1.365 + 0.045PC1 - 0.024PC2 + 0.093PC3$	2.030	1.030 - 1.080
C1-CT	0.668	$O_{3,t} = 16.136 - 0.016PC1 + 0.228PC2 + 0.032PC3$	1.987	1.024 - 1.132
C2-CT	0.579	$O_{3,t} = 16.975 + 0.018PC1 + 0.350PC2$	2.027	1.091

The results exhibited that the developed PCR models did not encountered any autocorrelation problem as the  $d$  values in between 1.961 (BG-NT-PCR) to 2.051 (BG-CT-PCR). All of the  $d$  values are above the upper limit ( $d_u$ ) 1.87 which clearly indicated that there are no autocorrelation in the developed models. The PCR models also did not encounter any multicollineary problems as the VIF values are significantly low in the range of 1.003 to 1.522. Low VIF values are expected in PCR models due to the usage of PCs as in the MLR models instead of the original variables. The capability of the PCR models in reducing multicollineary problems is widely known and have been utilized in air pollution studies (Lengyel et al., 2004; Abdul-Wahab et al., 2005; Gvozdić et al., 2011; Dominick et al., 2012).

Results suggested that during AT,  $\bar{R}^2$  around 0.6 was observed, with maximum value measured from UB-AT-PCR with 0.679 and minimum was 0.417 from BG-AT-PCR with an average value of 0.593. During AT, MLR consistently ranked the PC1 as the

most factor to O<sub>3</sub> variations followed by PC2, since PC1 represented a larger percentage of variance compared to PC2. PC1 was consistently loaded with variables such as the current O<sub>3</sub>, RH, T, UVB and WS, while CO, NO, NO<sub>2</sub> and SO<sub>2</sub> were loaded in PC2.

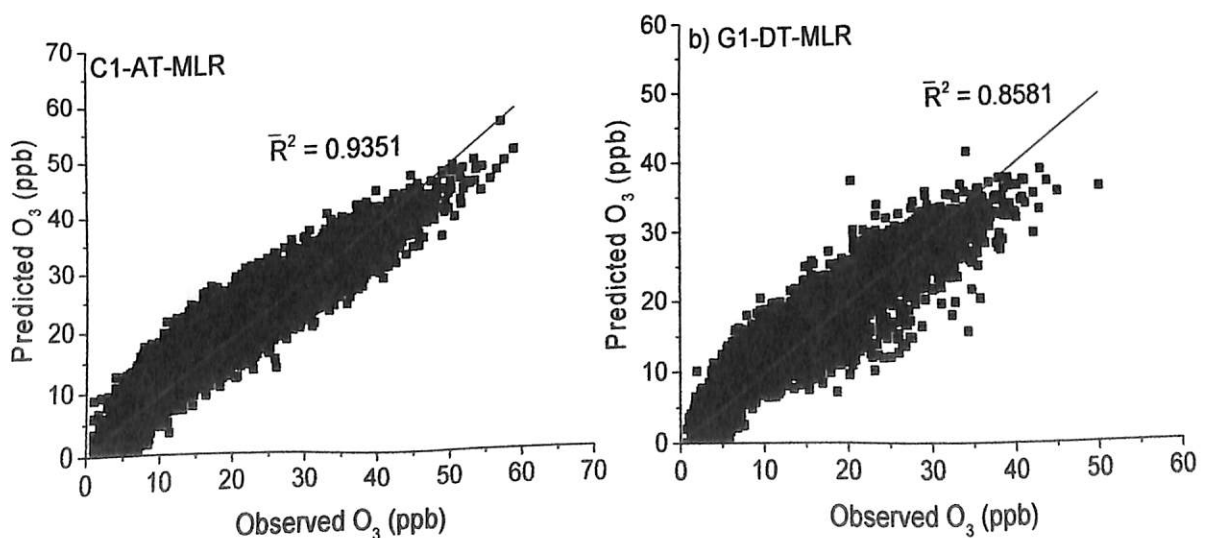
PCR models during DT also showed relatively lower  $\bar{R}^2$  values with an average of 0.445. Out of the nine models, G2-DT-PCR and C1-DT-PCR were the models that exhibited  $\bar{R}^2$  value higher than 0.5 with 0.556 and 0.564, respectively. Similar to the AT models, DT models also consistently ranked PC1 as the most important factor followed by PC2 and PC3. During this period, PC1 was mostly loaded with the current O<sub>3</sub>, RH, T, UVB and WS, while NO<sub>2</sub> and SO<sub>2</sub> was loaded in PC2. Whereas, CO and NO was loaded either in PC1 or PC2.

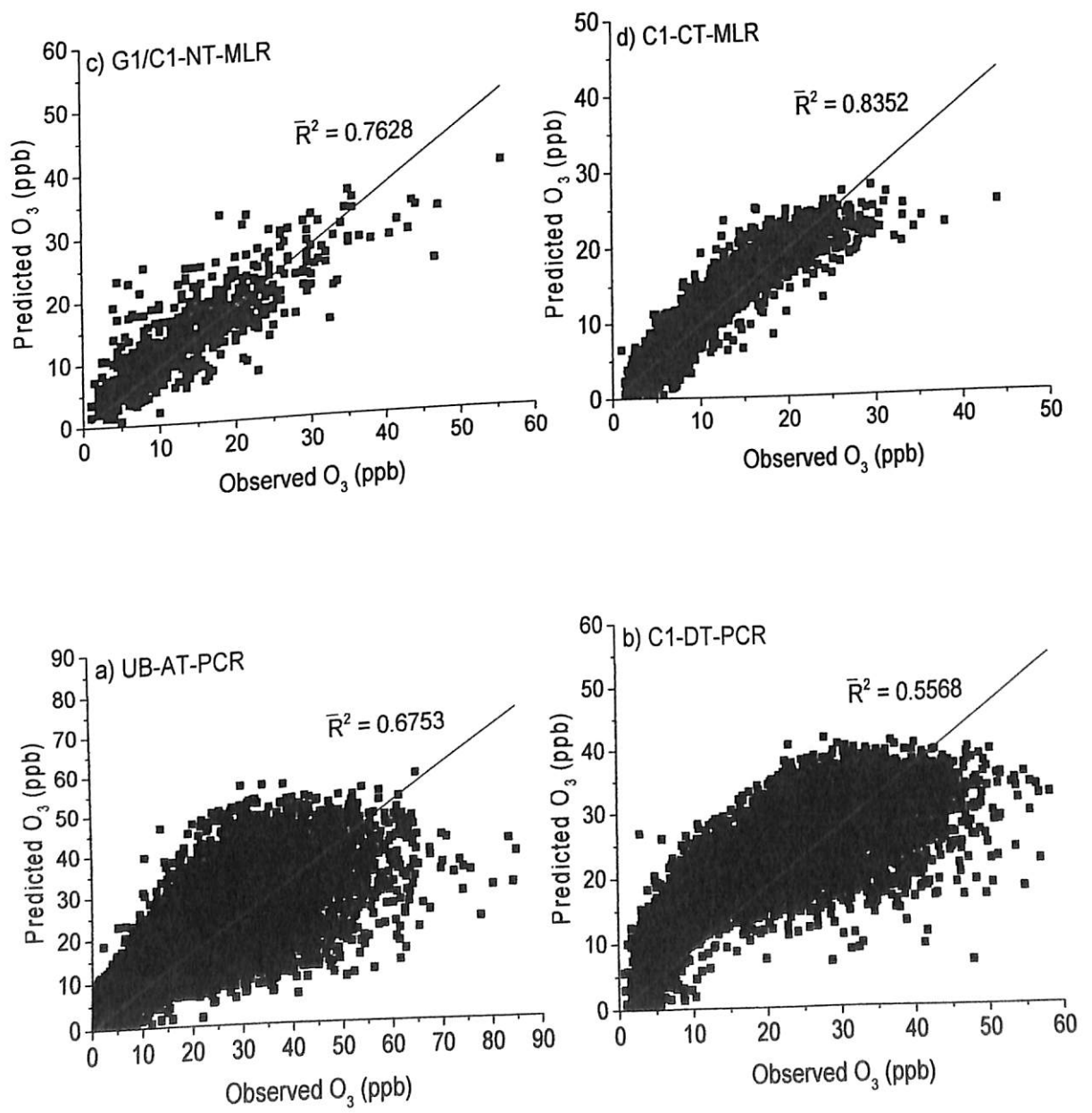
NT-PCR models showed significantly higher  $\bar{R}^2$  value compared to the AT and DT models. During this period, the average of  $\bar{R}^2$  was 0.607 with the maximum and minimum value 0.519 (UB-NT-PCR) and 0.657 (G2/C2-NT-PCR), respectively. The results also exhibited that there was an inconsistency in selecting the most important variable in predicting next hour O<sub>3</sub> concentrations during NT. For models such as BG-NT-PCR and G2/C2-NT-PCR, PC1 was selected as the most important variable followed by PC3 and PC2. Whereas for the rest of the NT-PCR models, PC2 is chosen as the most important factor followed by PC1. During NT, PC1 was mostly loaded with variables such as CO, NO, NO<sub>2</sub> and SO<sub>2</sub>, which are the precursors to O<sub>3</sub>. However, during NT, these variables were also responsible to O<sub>3</sub> destruction by various chemical reactions. Meanwhile, for most of the groups, PC2 was loaded with the current O<sub>3</sub>, RH and WS. The PC3 for BG-NT-PCR was loaded with RH, SO<sub>2</sub> and T, while for G2/C2-NT-MLR was loaded with NO<sub>2</sub> and T. The average  $\bar{R}^2$  value for PCR models during CT was 0.621, which is significantly higher than other time period. The maximum  $\bar{R}^2$  value for CT-PCR model was 0.783 which is measured for G2-CT-PCR models. However, SU-CT-PCR showed slightly lower  $\bar{R}^2$  value with 0.414 from three PCs. For CT-PCR models except for BG-CT-PCR, PC1 which was mostly loaded with CO, NO, RH, T, UVB and WS was

selected as the most significant variable followed by PC2 and PC3. Meanwhile, PC3 was ranked as the most important variable in BG-CT-PCR followed by PC2 and PC1. For this model, PC3 was loaded with SO<sub>2</sub>, PC2 with CO, NO<sub>2</sub> and current O<sub>3</sub> and PC1 was loaded with NO, RH, T, UVB and WS.

### The Best Prediction Model Based on Performance Indicators

The O<sub>3</sub> concentrations predicted using the optimal MLR and PCR models for each time period were plotted against the observed O<sub>3</sub> concentration are presented in Figure 24. The plots illustrated that there was a strong positive relationship between the observed and predicted O<sub>3</sub> concentrations especially during AT which recorded the  $\bar{R}^2$  up to 0.9351. Meanwhile, MLR models during DT and CT exhibited fairly strong positive relationship as their  $\bar{R}^2$  was 0.8581 and 0.8352, respectively. Despite the differences in the number of data used in developing these models, results showed that the performance of these models were quite similar. G1/C1-NT-MLR was the best NT-MLR model that produced a good relationship between the observed and predicted O<sub>3</sub> concentration as the  $\bar{R}^2$  was 0.7628.





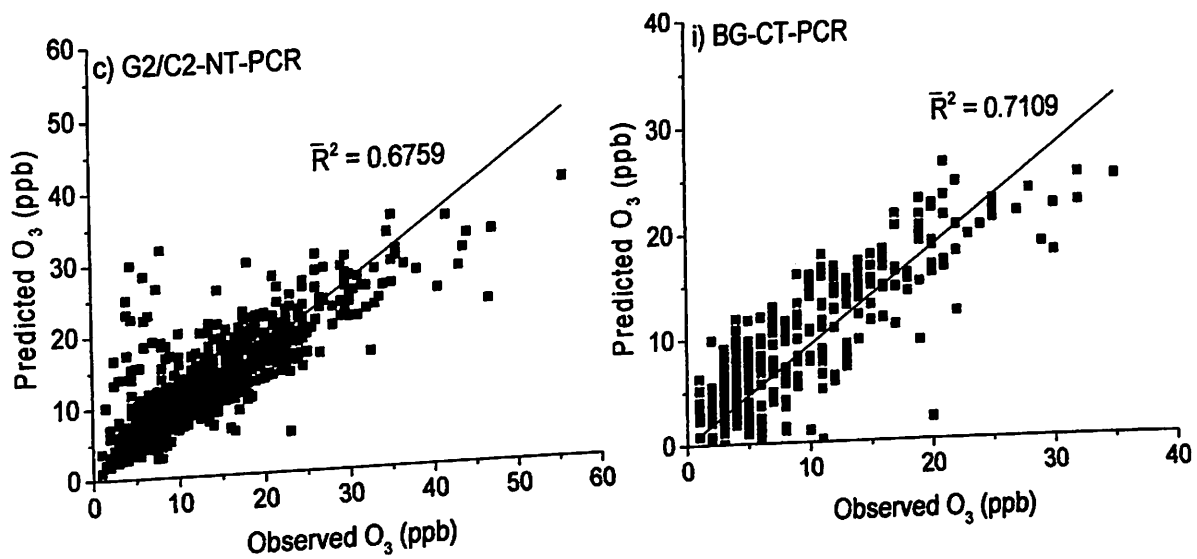


Figure 24 Scatter plot of observed and predicted O<sub>3</sub> concentrations using the best MLR and PCR models for different each time period

Meanwhile, the scatter plots of the observed and predicted O<sub>3</sub> concentrations using PCR models showed positive moderate correlation as their  $\bar{R}^2$  was from 0.5568 to 0.7109. Unlike the MLR models, the best PCR model was from CT period which BG-CT-PCR, while the C1-DT-PCR showed the weakest performance with  $\bar{R}^2$  of 0.5568.

C1-AT-MLR was selected as the best O<sub>3</sub> prediction model in this study. The model showed that the concentration of O<sub>3</sub> in Malaysia are really affected by the current O<sub>3</sub>, wind speed, NO, UVB, relative humidity, NO<sub>2</sub> and SO<sub>2</sub>. This finding was in line with the result presented by Ghazali et al. (2010) which stated that the current O<sub>3</sub> is a very important variables in predicting next hour O<sub>3</sub>. Nevertheless, studied by Abdul-Wahab et al. (2005) selected UVB, T, WS and NO as the variables that contribute to high O<sub>3</sub> concentration in Kuwait.

### Comparison between MLR and PCR Models



In this study, the usage of PCs as an input in MLR model was unable to improve the prediction performance of hourly O<sub>3</sub> concentrations. Table 13 shows the average of  $\bar{R}^2$  value for MLR and PCR models during different time period. The mean  $\bar{R}^2$  of PCR model was significantly lower than the MLR models with approximately around 31% for AT, 39% for DT, 10% for NT and 15% for CT. In addition, the values of the errors measures namely NAE, MAE and RMSE are smaller for MLR compared to PCR. The results of this study is in accordance with the findings presented by Lengyel et al. (2004) and Özbay et al. (2011) which revealed that MLR out-performed PCR models in modelling O<sub>3</sub> concentrations. Even though, PCR has more degree of freedom and offers variable combination in each PCs, the main purpose of using PCs as an input in MLR was to reduce multicollinearity problem among variables. However, based on the VIF value, the predictors used in MLR models did not encounter any multicollineary problems and the effect of multicollinearity was negligible. Thus, using PCA could not enhance the MLR performance.

Table 13 The average of  $\bar{R}^2$  value for MLR and PCR model

Time period	Adjusted coefficient of determination , $\bar{R}^2$		
	MLR	PCR	Differences (%)
AT	0.904	0.593	31.1
DT	0.833	0.445	38.8
NT	0.703	0.607	9.6
CT	0.774	0.621	15.3

In addition, Elbayoumi et al. (2014) mentioned that PCA is called an unsupervised dimension reduction methodology as reduction of the selected variables is applied without considering the correlation between dependent and independent variables. This might lead to prediction models loss of important information during the variable reduction stage. Result also illustrated that the rank of predictive power in several PCR models were not according to the order of PCs such as in IN-NT-PCR. PC1 which contained the information up to 38.19% was ranked as the second variable, while PC2 which was less explanatory was selected as the most important variable in predicting the O<sub>3</sub> concentrations.

## **OZONE MANAGEMENT AND PREVENTION FRAMEWORK IN MALAYSIA**

### **INTERNATIONAL OZONE MANAGEMENT: MONTREAL PROTOCOL**

The 1987 Montreal Protocol (MP) and its amendments established an ambitious schedule to reduce and eventually phase-out the production and consumption of chlorofluorocarbons (CFCs) and other ozone depleting substances (ODS). Following Malaysia's ratification in 1989, the government faced many challenges to implement its commitments given the importance of CFCs and Halons to the industrial sector (DOE, 2009)

The Government of Malaysia (GOM) created a National Steering Committee (NSC) to oversee the implementation of the national action plan, which paved the way for Malaysia's ratification in 1989. The NSC is comprised of a Technical Committee and industrial working groups (IWGs) on solvents, foam, aerosol, mobile air conditioning, refrigeration and fire protection. Other IWGs were established later to keep pace with the Protocol's amendments. In 1996, the Department of the Environment created the Ozone Protection Unit (OPU) to serve as the focal point and monitor Malaysia's phase-out activities.

The concept of integrated stakeholder partnership with the industrial sector is a key feature of Malaysia's response and implementation strategy. Partnerships with NGOs are also an integral part of the Government's awareness of the raising activities.

Although Malaysia fell under Article 5(1) of the Protocol, which extended a grace period of ten years to developing countries to meet international commitments, the government pursued a proactive strategy to reduce and limit the use of the controlled substances ahead of schedule.

Based on the work of the IWGs, Malaysia submitted its original country programme (CP) and the Action Plan to phase out ODS to the EXCOM in December 1991. The Plan aims to progressively reduce the consumption of ODS in each sector by 2000 through multiple means, including control measures (eg. enforcement of the Customs Duties Order), incentives (eg. exemption of import taxes on ODS recycling machines), partnerships for awareness activities, road shows and training, as well as project implementation and monitoring.

As a result of its proactive approach and financing through the Multilateral Fund, Malaysia has succeeded in phasing out more than 50% of CFCs and Halon in its manufacturing sector by 2000. It has thus met its obligations well in advance of the Protocol's timetable for Article 5(1) countries. Malaysia has also ratified the London, Copenhagen, Montreal and Beijing Amendments.

#### **LOCAL OZONE MANAGEMENT AND PREVENTION FRAMEWORK IN MALAYSIA**

The implementation of local air quality management (LAQM) scheme which incorporate the government bodies, the relevant bodies within the authority as well as the public will ensure the effectiveness of the implementation of LAQM. Local authorities have a major part to play in delivering cleaner air at the local level through the implementation of local ozone management and prevention (LOMP) scheme. However, the local authorities also need the support and cooperation from

the politicians, decision makers, government bodies, the relevant bodies within the authority as well as the public. This is to ensure the efficacy of the implementation of LOMP.

This study will develop the national and local framework of air quality management implementation in Malaysia. The structure of the developed framework (Figure 25) was derived from the current set up of interactions and line of command between federal government agencies, state government, local government and relevant bodies and agency.

The framework of local ozone management and prevention scheme proposed shown in Figure 5.1 (modified from Ramli et al., 2009). The proposed framework consists of three main stages:

Stage A enclosed by ( — ) is for LOMP policy development for haze preventions.

Stage B enclosed by ( ~~~~ ) is for preparations for carrying out LOMP processes

Stage C enclosed by ( .... ) is for LOMP implementation processes

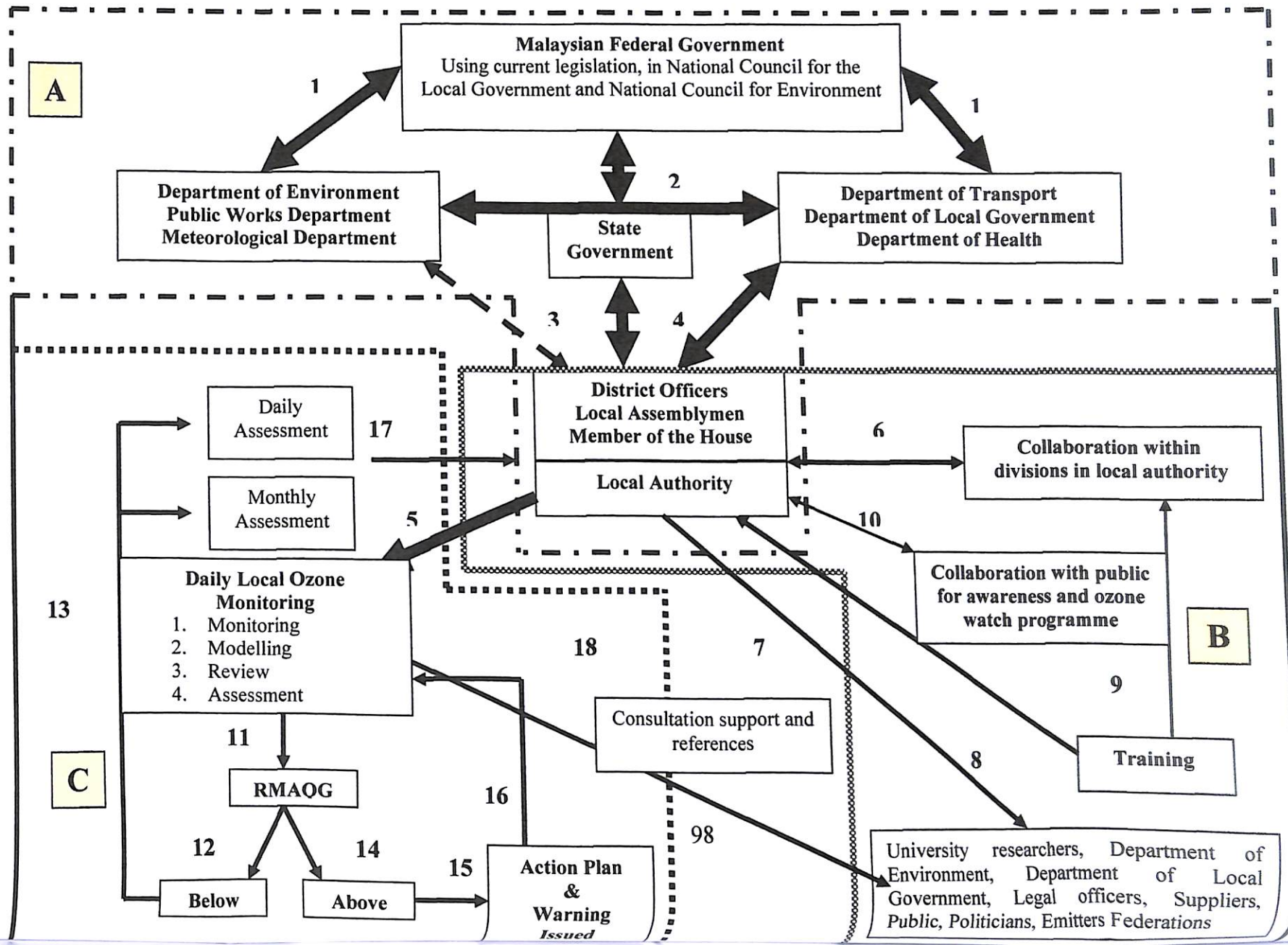
The arrows and numbers show the relationships between one step to another in the framework. The thickness of the arrows shows the binding strength of responsibility with the leads implying direction of communication.

### **Local Ozone Management and Prevention Policy Development**

Stage A of the local air quality management framework is local air quality management policy development, which involves the collaboration between the Federal Government Councils, State Government, State Assemblymen, Parliament Member, Department of Environment, Department of Local Government, Department of Health, Department of Transport, Meteorological Department, District Officers and Local Authorities. In the current air quality management framework, the Federal Government, through the Department of Environment, manages all the

air quality management activities in Malaysia. However, through this new integrated ozone management and prevention schemes, its function will be discharged through the State Government and Assemblyman, Department of Local Government, district officer, local authority, as well as all related federal agencies. Figure 5.1 show the proposed framework of local ozone management and prevention to be adopted by the authorities in Malaysia (modified from Ramli et al., 2009).

Figure 25: Proposed integrated local ozone management and prevention scheme to reduce the risk of exposure to secondary pollutants in Malaysia



The Federal Government has the executive power in the country. Therefore, the policy to implement local ozone management and prevention scheme should be initiated by the Federal Government of Malaysia under the National Council for the Local Government and National Environment Quality Council platform. The framework shows that the Department of Local Government and Department of Environment are the main federal agencies involved in local air quality management. In addition the Public Works Department, Department of Health, Meteorological Department, and Department of Transport may have direct and indirect influence on the policy made under the proposed scheme. The Department of Local Government, who manages local authority affairs in Malaysia, is administered under the legislation of National Council for Local Government at federal level. It is the local authorities who will be consulted in implementing any development project and environmental management at local level. Due to this reason, the National Council for Local Government will delegate power to the Department of Local Government [represented by thick arrow (1)]. The Department of Local Government will be the Federal Government representative in charge of the matters that are involved with the local air quality management. With the power given by the National Council for Local Government, the Department of Local Government could issue directive to the District Officer to carry out the local air quality management [represented by thick arrow (4)]

The involvement of the Department of Environment is important because of their experience and expertise in managing air quality. Therefore the National Council for Local Government and National Environmental Quality Council will request the Department of Environment [represented by thick arrow (1)] to cooperate with the Department of Local Government. Both of the departments have the expertise in two main aspects in local air quality management which are, air quality management and local authority. The cooperation between these two departments is highly recommended. So that, sufficient and effective ways for the Department of Local Government to manage the local ozone management and prevention matters will be produced. The consultation needs to be done to achieve consensus between them. To achieve the consensus, it is important for them to study and truly understand the responsibilities and legislations of both departments. The legislations that need to be explored to extent are Environmental Quality Act 1974 and Local Government Act

1976. Nevertheless, interactions, coordination and cooperation between all relevant federal agencies and state agencies are represented by thick cross arrow (2).

The involvement of State Governments is important since it coordinates and manages all state activities. The Federal Government will consult with the State Government about the implementation of local air quality management [represented by thick arrow (2)]. The State Governments could observe and issue directive to the District Officer to ensure local air quality management is carried out [represented by thick arrow (2)]. The State Government also has to be in touch with the Federal Government in order to ascertain the actions taken by the local authorities in implementing local air quality management is in accordance with Federal Government requirements.

The Department of Environment and local authorities which includes the District Officer have existing link [represented by dotted arrow (3)] under Order 2, Schedule (d) – (i) Environmental Quality (Regulation of Powers) (Investigations of Open Burning) Order 2000. This is the only existing official arrangement that link the Department of Environment and the local authorities with regards to air pollution control at local level under the Environmental Quality Act, 1974. No specific legislations pertaining to ambient concentration of ozone are available. Hence perhaps area with monthly mean concentrations of daytime ground level ozone should have special actions as specified in Stage C.

Local government comprises two levels, district administration and local authority. The district officer heads the District Council and is responsible for the development of the district as a whole. Coordination of the development activities, including local air quality management implementation, although it is done through various committees, but most of which chaired by the district officer. There are two types of local governments, municipality for large towns, and district council for small urban centres. In implementation of local air quality management, action is being undertaken primarily by local authority, which is directly responsible and accountable for its actions. It will implement all the air quality management activities on their own, in accordance with the government legislations. Since ozone



are secondary pollutants, the LOMP should focus on reducing the precursors of ozone.

### **Preparation for Carrying Out Local Ozone Management and Prevention Scheme**

Stage B of the local air quality management framework is preparations for carrying out air quality management processes. University researchers, Department of Environment officers, Department of Local Government officers, legal officers, the suppliers, general public, politicians, Emitters federations should work together in a group to prepare technical guidelines for the use of local authorities in implementing LOMP [represented by medium arrow (7)]. The technical guidelines should contain most of the aspects in local air quality management such as monitoring, modelling as well as appropriate ways to do review and assessment on air pollution. This should be followed by training regime and collaborations as depicted by arrow 6 to 10.

The local authority should build an effective link with all relevant bodies within the authority, such as Health Authorities, Public Works Department, Town Planners, Transport Authority as well as Manufacturer Federations [represented by medium arrow (7)]. This is needed since these relevant bodies have important roles in the local area. It is important to the local authority to communicate with the Public Works Department, Town Planners, Transport Authority as well as Manufacturer Federations in the area to make sure there will not be too many developments simultaneously that will cause air quality to get worse. The consensus should be achieved between the local authority and these relevant bodies including the politicians and counsellors in order to ascertain the effectiveness of LOMP management implementation in their area. The willingness to cooperate will help to improve the local air quality at their local area.

This stage also emphasises on the preparation of the local authority personnel to implement LOMP [represented by medium arrow (9)]. Having the in-house personnel can avoid the local authority to pay money for consultants to do the air quality management work for them. However, before this could happen, the local authority personnel, who are very qualified to do the work, should be trained first.

The panel of experts on LOMP, who are responsible to prepare the technical guidelines for local air quality management, will carry out the training. Training is an important step in the preparation process of local air quality management implementation because through the training provided, the local personnel can learn and gain knowledge about air quality management. Air quality management especially LOMP is alien to the local authorities since the local authorities in Malaysia only manage solid waste disposal. Therefore, it is important for the local authority personnel who will be in charge of the LOMP to attend the training session in order to provide the most appropriate action to be taken in their area. Adequate skill must be developed to carry out the LOMP and air quality management at the end of the training session.

The personnel chosen to implement the LOMP is not only limited to officers from local authority, but also personnel from the relevant bodies within the authority. Training should also include the personnel from the relevant bodies within the authority [represented by thin arrow (9 and 10)]. The local authority personnel, of course, are capable of determining this standard following training since they know the real state of their area in greater detail. The personnel from other relevant bodies must also have known the current situation in the local area although in a particular matter. As an example, the manufacturer federation personnel must have known the types of pollutants that are polluting the air produced by the industrial processes. The same situation is true for the transport authority since the Department of Transport personnel will know the number of vehicles moving around the area. With the information they have, they can help the local authority to find solutions in preventing these sources; emissions from industries and vehicles. Therefore, the personnel from the relevant bodies within the authority are suitable candidates to attend the training session that will be carried out by a panel of experts.

Other than collaborating with the relevant bodies within the authority, it is also important for local authority to gain support from the public [represented by thin arrow (10)]. The local authority should let the public know about the LOMP implementation since they are the local people who are being effected by the pollution. They are should be informed about the current air pollution problem and the benefit that can be achieved through the implementation of this new air quality

management scheme in improving air quality. The public can then put pressure on the politicians to make sure that the local air quality management can be implemented successfully. The feedbacks and opinions from the public can also help the local authority to develop better actions to be taken in their local air quality management scheme.

Stage C is the implementation stage whereby local authority plays a vital role in monitoring daily ozone concentrations as required by LOMP. Daily monitoring, modelling, review and assessment are carried out by NGO volunteers and local authority personnel [thick arrow 5, medium arrow 11 to 17]. Daily assessment and monthly assessment were made based on Malaysian recommended air quality guideline (RMAQG) for ozone. Action taken will depend on either below or exceeding the RMAQG. Action plan to reduce ozone concentrations as well as issuance of warning to local public are continuous monitoring benchmarking to ensure lower ambient ozone. Finally, online helpdesk for support and reference should be made available from relevant entities to ensure high quality ozone monitoring campaign and results [arrow 18].

### **Conclusion**

From this research, it can be concluded that the O<sub>3</sub> concentrations in all stations were relatively high with daily maximum of O<sub>3</sub> concentration surpassing 100 ppb for most of the stations except for Kota Bharu, Kota Kinabalu, Jerantut and Kuching. The highest O<sub>3</sub> concentration was 175 ppb in Kajang which was recorded in June 2000. The time series plot of the daily maximum O<sub>3</sub> concentrations revealed that O<sub>3</sub> concentrations in Shah Alam, Kajang, Gombak, Klang, Nilai and Bakar Arang frequently showed exceedances cases. The total number of exceedances in between 1999 to 2010 was 1995 hours or 166.25 hour/year. The highest number of exceedances was measured in 2004 with 315 hours and 93.5% of the total exceedance was contributed by the urban stations such as Shah Alam (885 hours) and Kajang (551 hours).

In Malaysia, O<sub>3</sub> exhibited typical diurnal uni-modal trend characterized by high daytime concentrations and low nighttime concentrations. The minimum O<sub>3</sub> concentrations were constantly measured at 8 a.m. except for Gombak and Kota Kinabalu which was measured at 7 a.m. Meanwhile, the maximum O<sub>3</sub> concentration was measured from 12 p.m. to 4 p.m., with the majority measured at 2 p.m.. O<sub>3</sub> will significantly be lower during November to January each year, attributed by a higher rate of wet deposition during the North East Monsoon. O<sub>3</sub> maps revealed that the O<sub>3</sub> pollution was more prominent in the west coast of Peninsular Malaysia as the level of O<sub>3</sub> precursors being significantly higher due to high anthropogenic activities and abatement efforts should be more focused in this region than any other places in Malaysia.

Next, the results from PCA confirmed that NO<sub>2</sub>, NO, T and UVB were the most important factors that controlled the O<sub>3</sub> transformations and fluctuations as the percentage of total variance explained in between 60 % to 80 % was recorded. Even though, PCA gave inconsistent results to select the most important factor through PC1, the composite diurnal plots of O<sub>3</sub>, NO<sub>2</sub>, NO, T and UVB clearly indicated that O<sub>3</sub> formation was more influenced by NO<sub>2</sub> and NO concentrations than T and UVB. The CCP of O<sub>3</sub> production is also being introduced in this study. The time where CCP occurs was determined from the composite diurnal plots as the interception point between O<sub>3</sub>, NO<sub>2</sub> and NO line by which in all stations measured at around 9 a.m. or 10 a.m.. Stations such as Shah Alam, Kajang, Gombak, Nilai, Perai, Klang, Johor Bahru, Seberang Jaya and Pasir Gudang recorded average time for CCP is ±10 a.m.. Meanwhile, time for CCP at Bakar Arang, Kemaman, Bukit Rambai, Ipoh, Taiping, Kota Bharu, Jerantut and Kuching is ±9 a.m.. Kota Kinabalu is the sole station that recorded time for CCP at ±8 a.m.. CCP were also determined by calculating the differences between rate of NO<sub>2</sub> photolysis and NO titration and results showed that CCP usually occurred between 8 a.m. to 11 a.m.. Based on CCP, the critical conversion time (CT) used was during 8 a.m. to 11 a.m.. The existence of CCP at selected stations were verified by collecting new monitoring records in Bakar Arang (sub urban), Nilai (industrial) and Shah Alam (urban). Albeit, there was an hour difference between CCP and verified CCP in Bakar Arang and Nilai, the points were still between 8 a.m. to 11 a.m..

The next hour O<sub>3</sub> prediction models were developed using MLR and PCR during AT, DT, NT and CT for different group of monitoring stations based on DoE classification, ranking of means and cluster analysis. MLR prediction models which are a simpler model produced approximately 23% higher  $\bar{R}^2$  than PCR models and the best O<sub>3</sub> prediction model in this study was C1-AT-MLR which was also one of the MLR model. This model showed the highest prediction accuracy indicated by the value of  $\bar{R}^2$  (0.9351), PA (0.9671) and IA (0.9831) and least prediction errors. The results also proven that the developed O<sub>3</sub> prediction models using new group of monitoring stations produced higher performance in terms of accuracy and error measures than the DoE group due to similar range in O<sub>3</sub> concentrations used in the groups. Even though, the O<sub>3</sub> prediction models during CT was developed using smaller dataset, the capability of these models to predict the O<sub>3</sub> concentration was relatively good as the average  $\bar{R}^2$  for CT-MLR is 0.7881 and CT-PCR is 0.5800. The applicability of the prediction models to predict the next hour O<sub>3</sub> concentrations in urban, sub-urban and industrial areas were verified as the models showed moderate and good correlation between the observed and predicted O<sub>3</sub> concentrations.

The introduction of CCP through this study is proven to be useful in obtaining more information on ground level O<sub>3</sub> transformation from its precursor behaviour. Identification of CCP may provide some basis in term time period for policy development and enforcement in order to properly control O<sub>3</sub> air pollution in ambient air. It is believed that O<sub>3</sub> accumulation process will be disturbed if the level of its precursors being reduced during the critical conversion time thus lead to O<sub>3</sub> reduction. Whereas, if high level of O<sub>3</sub> precursors is continued being emitted during CCP, O<sub>3</sub> accumulation will be promoted, thus lead to increment in O<sub>3</sub> concentration. This study has provide some fundamental information regarding the CCP, it provide positive indication of its usage in managing O<sub>3</sub> problems and create the possibilities to be further explored.

Local ground level ozone should be implemented soon in Malaysia to control exposure of the public.

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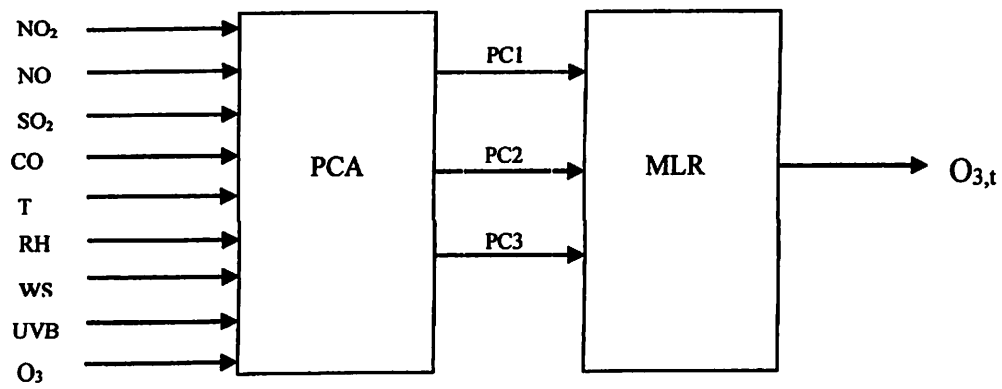


Figure 7 Architecture of a PCR models using PCA outputs as the input to MLR

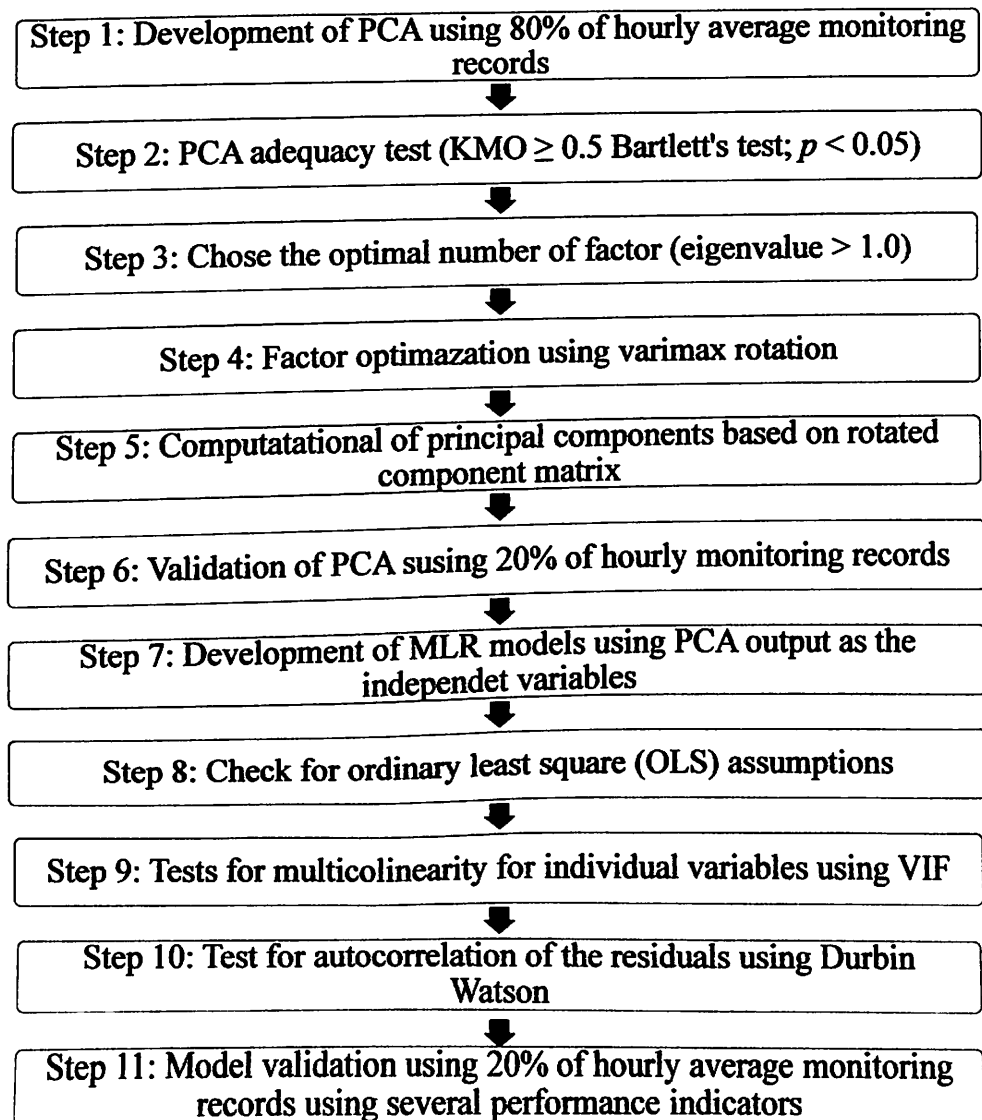


Figure 8 Procedure of developing PCR models during the predetermined time range

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**ASSESSMENT OF FLUCTUATIONAL AND CRITICAL  
TRANSFORMATIONAL BEHAVIOUR OF GROUND LEVEL OZONE**

**by**

**NORRIMI ROSAIDA BT AWANG**

**Thesis submitted in fulfillment of the requirements  
for the degree of  
Doctor of Philosophy**

**August 2015**

**STUDY ON THE FLUCTUATION BEHAVIOUR  
AND RELATIONSHIP OF O<sub>3</sub> AND PM<sub>10</sub> IN  
PENINSULAR MALAYSIA AND BORNEO**

**By**

**SITI AMINAH BTE MD SAAD**

This dissertation is submitted to

**UNIVERSITI SAINS MALAYSIA**

As partial fulfillment of requirements for the degree of

**MASTER OF SCIENCE  
(ENVIRONMENTAL ENGINEERING)**

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**INFLUENCE OF METEOROLOGICAL  
FACTORS ON THE FORMATION OF OZONE  
FROM NITROGEN DIOXIDE**

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