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Variation of Surface Ozone Recorded at the Eastern Coastal Region of the Malaysian Peninsula

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Abstract: Problem statement: Variations of ozone (O_3) concentrations in the Eastern Coastal Region of the Malaysia peninsula were investigated using data obtained from the Malaysian Department of the Environment. The aim of this study was to determine the monthly and seasonal variations of ozone concentrations at all monitoring sites. This study deals with the air quality data recorded at four air quality monitoring stations in the East Coast of the Malaysian peninsula over a ten year period (1997-2006). **Approach:** We focused on the usage of S-Plus and SPSS to analyze this data. The S-Plus programming was used to impute missing data and SPSS was used to obtain the variations of ozone and also to clarify the relationship between stations. **Results:** Over the entire 10 year period (1997-2006), the trend in annual baseline ozone generally increased each year for all the four monitoring sites. There was also a seasonal variability in the measured ozone levels with high concentrations during the southwest monsoon and the northeast monsoon season, producing a significant increase in the amplitude of the seasonal cycle. The results also shown that the highest ozone concentrations were recorded at the Bukit Kuang air monitoring station (S1), with a daily mean value of 19 ppb followed by the Indera Mahkota air monitoring station (S2). The concentration of ozone recorded at Kota Bharu (S3) and Kuala Terengganu (S4), two stations located in the city centre, was found to be lower than the values recorded at Bukit Kuang and Indera Mahkota. The correlation between O_3 and NO is high at Kuala Terengganu (S4) ($\rho = -0.579$), whilst the relationship between O_3 and NO_2 are high ($\rho = -0.397$) at Indera Mahkota (S2). **Conclusion:** The concentration of ozone in the East Coast of Malaysian peninsula depends on the concentration of NOx and seasonal meteorological factors.

Key words: Concentration of ozone, Time series, Oxides of nitrogen, The Malaysian peninsula, Total Hydrocarbon (THC), Flame Ion Detector (FID), Package for Social Science (SPSS), ozone concentrations, photochemical production, solar radiation

INTRODUCTION

Photochemical oxidants are trace species, which are formed during the atmospheric photo-oxidation of a variety of trace gases (Kley *et al*., 1998; Rajab *et al*., 2010). Ozone, one of the most important photo-oxidants in the urban environment, originates from the in-situ photochemical production in the reactions of its precursors (NOx, CO, VOCs) and from vertical and horizontal transport (Minoura 1999; Latha and Badarinath, 2004; Badarinath *et al*., 2007; Lin *et al*., 2008; Ahmad *et al*., 2010). Due to the nature of ozone, its photolysis in the troposphere has been shown to be directly related to ultraviolet solar radiation at a wavelength of around 300 nm, followed by a reaction of OH radicals, which take part in reactions responsible for the oxidation of other gases present in the atmosphere (Dentener and Crutzen 1993; Atkinson, 1997; Guicherit and Roemer 2000; Muzathik *et al*., 2010; Cansee *et al*., 2010; Charoensawan and Wannagosit, 2010).

The effects of ozone were introduced by Middleton *et al*. (1950) over 50 years ago (Lehman *et al*., 2004; Ali *et al*., 2009; 2010). On the scale of 100 years, the increasing trends of tropospheric ozone are qualitatively in agreement with emissions trends of precursors. This has led to the increase in the

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tropospheric ozone level becoming one of the most crucial environmental problems needing to be solved in the coming decades due to its adverse effects, particularly on vegetation (Finlayson-Pits and Pitts 1997; Ishii *et al*., 2004; Ishii *et al*., 2007; Shan and Yin, 2008; Soleimanzadeh *et al*., 2010; Zouzoulas and Koutroubas, 2009). Hourly mean ozone recorded by the European Monitoring and Evaluation Programme (EMEP) rural ozone monitoring network, rarely approach zero and typically are 2-5 ppb in relatively clean air masses which have had little influence from human activity (Hjellbrekke and Solberg, 2005; Azzi and Duc, 2008).

Recent investigations indicate that the concentration of ozone in the Earth's atmosphere is changing. Although there is a good agreement regarding a rise in background levels over the past century, in recent decades divergent trends in tropospheric ozone have been observed over different regions of the globe, especially in the north hemisphere (Vingarzan 2004; Zouzoulas and Koutroubas, 2009). Several researchers have demonstrated through data, the existence of a correlation between seasonal trends in the level of ozone concentrations are the local surroundings of various monitoring stations (Adeeb and Shooter 2004; Duenas *et al*., 2004; Feng *et al*., 2005; Lee *et al*., 2009; Piikki and Klingberg, 2009; Azmi *et al*., 2010). The trend analysis of ozone always has maintained a relationship with the trends of VOC, NO and NOx in both urban and suburban areas (Atkinson, 2000; Ghazali *et al*., 2010; Keuken *et al*., 2009; Martin *et al*., 2009; Milt *et al*., 2009). Local concentrations of ozone are reduced in the vicinity of heavy vehicular traffic as a results of ozone scavenging by NO and NO_2 (Atkinson, 2000; Ahammed *et al*., 2006; Tu *et al*., 2007; Chen *et al*., 2009). On the other hand, areas with less traffic, specifically downwind of a monitoring station, may have a higher ozone concentration due to active photochemistry in an air mass enriched with precursor chemicals from motor vehicle exhaust gases (Lippmann, 1991; Afroz *et al*., 2005). In the case of Southeast Asia and other tropical countries, biomass burning especially from forest fire is also expected to contribute to the amount of ozone in the atmosphere. This is in addition to non-methane hydrocarbons from tropical forests (Komala and Saraspriya, 1996; Liu *et al*., 1999; Chan and Chan, 2000; Pochanart and Kreasuwun , 2001; Jusuf and Ibrahim, 2009).

The approach taken in this study is to focus on the surface ozone time series with relatively long records (10 years) and locations which are representative of the East Coast of Malaysia's geographical regions. This study also aims to quantify the overall monthly and daily trend for each of the monitoring sites other than the influence of atmospheric pollutants related to the ozone concentration recorded at each station. This study uses continuous observation for 10 years with data recorded every hour.

METERIALS AND METHODS

Sampling sites: The Eastern Coastal Region of the Malaysian peninsula (Fig. 1) has been chosen as the study site because it allows for the air quality in both urban and suburban areas to be determined, rather than solely the urban area as would have been the case had Kuala Lumpur been selected as the site of study. Furthermore, the East Coast of the Malaysian peninsula is a low pollutant area and was chosen specifically so that air quality could be determined in such a region. The meteorological condition in this region influence by the South West monsoon from June to September and North East monsoon November to March. The inter monsoon seasons usually occurred in month of April and October. The term East Coastal region is particularly used in Malaysia to refer to any one of the three states in the Malaysian peninsula facing the South China Sea. These states are Pahang, Kelantan and Terengganu, which have a total area of 64,911 km^2, a total population of 4.5 million and a total density of 215 people km^{-2}. The description of these monitoring stations is presented in Table 1.

The Bukit Kuang air monitoring station (4.16° N, 103.25°E; S1) located at a primary school and considered to be as far from urban areas but in close proximity to the main road connecting Kemaman and Kuala Terengganu. It is also around five kilometers from the Teluk Kalung Industrial areas. The Indera Mahkota air monitoring station (3.49°N, 103.17°E; S2) located in Bandar Indera Mahkota Primary School is situated in a residential area about 4 kilometers northeast of Kuantan Town. This station is located in close proximity to the main road of the East Coast Expressway (LPT) via the Kuantan Interchange. Bandar Indera Mahkota is also more accessible to Sultan Haji Ahmad Shah Airport and Gambang through the Kuantan Bypass Highway. The Kota Bharu air monitoring station (6.09°N, 102.15°E; S3) is located at Sultan Ismail College in Kota Bharu city center (capital state of Kelantan). There is quite a lot of traffic in this area, particularly during the morning and late afternoon rush hour. The Kuala Kuala Terengganu (5.18°N, 103.07°E; S4) air monitoring station is situated at the Chabang Tiga Primary School, which is located near to the Kuala Terengganu city center.

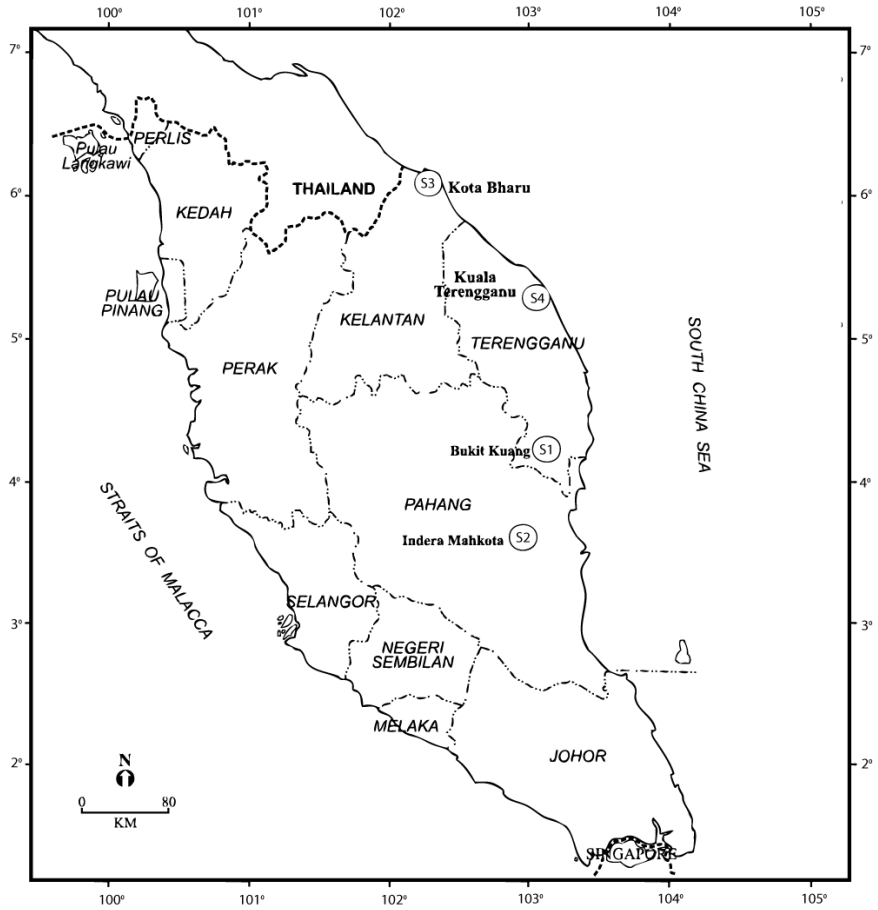

Fig. 1: Map of the Peninsula Malaysia with location of monitoring stations in the east coast of Malaysian peninsula

Table 1: Location and description of selected air monitoring stations on the eastern coast of the Malaysian peninsula

Air monitoring station	Location	Background	Latitude	Longitude
S1	Bukit Kuang Primary School, Kemaman, Terengganu	Rural	4.16°N	103.25°E
S2	Indera Mahkota Primary School, Indera Mahkota, Pahang	Semi-Urban	3.49°N	103.17°E
S3	Sultan Ismail College, Kota Bharu, Kelantan	Urban	6.09°N	102.15°E
S4	Chabang Tiga Primary School, Kuala Terengganu, Terengganu	Urban	5.18°N	103.07°E

For this reason it also affected by busy traffic, particularly during the rush hour in the morning and late afternoon.

Ozone and other parameters data collections: The data for our investigation was obtained from the air quality monitoring sites in Malaysia, which though owned by the Department of the Environment (DOE) are managed a private company, Alam Sekitar Sdn. Bhd (ASMA). This data contains the monthly hourly average of ozone concentrations, oxides of nitrogen (NO and NO_2), Total Hydrocarbon (THC) and hourly meteorology measurements for variables such as: wind speed, air temperature, relative humidity and ultraviolet solar radiation.

The ozone concentration at Alam Sekitar Sdn. Bhd stations were measured using Teledyne Ozone Analyzer Model 400E UV Absorbtion. The analyzer uses a system based on the Beer-Lambert law for measuring low ranges of ozone in ambient air. The concentration of nitrogen oxides were determined using chemiluminescence measurement principle, coupled with state-of-the-art microprocessor technology for monitoring high and medium levels of nitrogen oxides (Teledyne Models 200EH and 200EM) while the concentration of total hydrocarbon THC was determined using field proven Flame Ion Detector (FID) (Teledyne Model 4020). In addition, meteorological parameters such as, wind speed, UV radiation and humidity also were also recorded at each station.

Missing data: The "nearest neighbor method" has been used to impute missing data. This method for imputation is considered to be the simplest scheme available in the S-PLUS FinMetrics module, in that the endpoints of the gaps are used as estimates for all the missing values (Eq. (1)).

$y = y_1$ if $x \leq x_1 + (x_2 - x_1)/2$,
$y = y_2$ if $x > x_1 + (x_2 - x_1)/2$,

Where:
Y = The interpolant
X = Time point of the interpolant
y_1 and x_1 = The coordinates of the starting point of the gap
y_2 and x_2 = The coordinates of the end point of the gap

The data that was missing will be interpolated through the nearest neighbour value available using S-PLUS (Junninen and Niska, 2004; Ali *et al*., 2008).

Statistical analysis: The data for ozone concentrations at each monitoring site was analyzed using the Statistical Package for Social Science (SPSS) 14.0 software, which was used to obtain the variations of ozone and also to clarify the relationship between stations. The statistics included in this software, which analyze the ozone concentration data, are descriptive statistics (frequency, graph and box plot) and bivariate statistics (correlation matrix). To see the relationship between different air pollutants at each station, the Spearman correlation was conducted in this study.

RESULTS

Variations in baseline ozone mixing ratios from 1997-2006: In general, average annual concentrations of ozone were recorded at the highest concentration in Bukit Kuang (S1) followed by Indera Mahkota (S2), Kota Bharu (S3) and Kuala Terengganu (S4). The annual mean of ozone concentrations recorded at Bukit Kuang (S1) increased steadily from 17 ppb in 1997 to 20 ppb in 2003, followed by a small decline and subsequent stabilization (Table 2 and Fig. 2). For Indera Mahkota (S2), the trends for ozone fluctuated between 1997 and 2003 and then became constant at the highest concentrations during 2004-2006 at 18 ppb ozone concentrations. The opposite occurred with Kota Bharu (S3), where the constant annual mean of ozone was recorded at the low concentrations of 12 ppb in 1998-2003 and then slightly increased to 14 ppb in 2004-2006. The Kuala Terengganu (S4) annual mean concentrations have the highest value at 17 ppb in 1999 and then decreased until 2003 before the value increased and returned to 17 ppb in 2004.

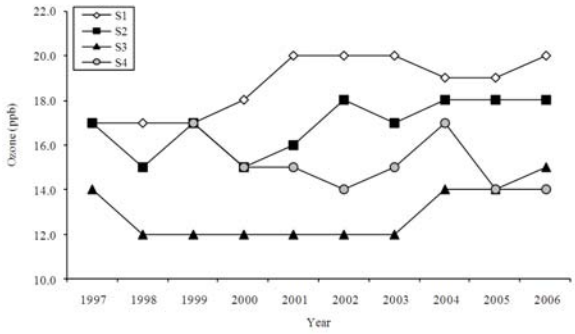

Fig. 2: Anual ozone concentrations recorded at selected monitoring stations

Monthly mean baseline levels of ozone at Bukit Kuang (S1) reached a peak of 27 ppb in July 2003, 25 ppb in August 2004 at Indera Mahkota (S2), 19 ppb in May 2005 at Kota Bharu (S3) and 23 ppb in December 2003 at Kuala Terengganu (S4). However, the month that shows the lowest range of ozone at Bukit Kuang (S1) was October 1999 with 11 ppb. This value has the same concentration as Indera Mahkota (S2) in November 2000 and 2003. The sites with the lowest concentrations of ozone among all of the four monitoring sites is Kota Bharu (S3) (Fig. 3), where the lowest concentration of 7 ppb was recorded during October 2000 and 2001. The month which shows the lowest concentration of ozone at Kuala Terengganu (S4) is October with 9 ppb.

Monthly monitoring: Figure 3 shown the boxplot of monthly ozone concentration. The boxplot indicate that the concentration of ozone is higher during May to September (southwest Monsoon) and December to March (northeast Monsoon) and recorded at the lowest concentration during the inter-monsoon season (April and October).

Correlation between ozone and its precursors (NOx and THC) and meteorological factors: The concentration of ozone at all sampling stations has a negative correlation with the concentrations of NO, NO_2 and Total Hydrocarbon (THC) (Table 3). No THC concentration was observed at Kuala Terengganu (S4) as there was no instrument with which to measure THC concentration at this site. The results from Fig. 4 show that the monitoring stations with high concentrations of NO, such as Kota Bharu (S3) and Kuala Terengganu (S4), recorded low concentrations of ozone in comparison to Bukit Kuang (S1) and Indera Mahkota (S2). The strength of a linear relationship between ozone and NO is high at Kuala Terengganu (S4) based on the

Table 2: Monthly mean of ozone concentration (ppb) at four sites on the east coast region of Malaysian peninsula

	Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Mean
S1	1997	24	16	17	16	19	22	22	26	26	20	16	17	20
	1998	16	14	19	20	16	17	18	15	20	17	14	15	17
	1999	17	20	12	14	18	14	22	22	21	11	13	22	17
	2000	19	20	13	15	21	16	17	22	20	18	14	17	18
	2001	17	19	18	17	21	21	26	23	23	17	19	21	20
	2002	22	19	16	15	20	24	25	25	23	19	15	15	20
	2003	22	16	15	13	20	21	27	23	24	19	14	26	20
	2004	20	19	17	15	21	21	18	26	19	18	14	19	19
	2005	24	14	18	15	21	21	21	24	21	15	15	17	19
	2006	21	16	15	15	17	21	23	25	24	20	16	23	20
S2	1997	19	14	15	15	18	19	18	21	23	16	12	12	17
	1998	14	12	14	16	16	17	19	15	18	16	15	14	15
	1999	14	16	13	16	17	20	22	22	24	13	12	16	17
	2000	13	17	13	15	18	16	19	18	17	14	11	14	15
	2001	12	16	14	14	15	16	20	16	17	16	15	19	16
	2002	19	18	18	16	18	20	18	21	20	22	14	13	18
	2003	19	15	16	14	19	20	16	21	19	15	11	19	17
	2004	16	18	16	17	20	20	17	25	18	19	15	18	18
	2005	23	18	19	17	19	19	19	22	20	14	14	16	18
	2006	20	14	17	15	16	18	19	22	22	19	13	18	18
S3	1997	12	12	15	14	15	19	17	15	16	12	11	13	14
	1998	12	10	16	17	15	14	12	11	9	8	9	12	12
	1999	14	13	11	10	13	14	13	12	11	9	9	17	12
	2000	14	15	10	12	12	11	13	10	12	7	11	11	12
	2001	12	14	11	11	14	12	16	9	11	7	12	13	12
	2002	15	13	12	11	14	15	13	13	9	9	8	9	12
	2003	15	10	10	9	11	18	11	13	11	8	10	18	12
	2004	16	16	17	16	17	15	13	16	14	10	8	8	14
	2005	13	9	11	16	19	17	16	16	14	12	13	15	14
	2006	16	13	15	14	15	15	18	16	15	14	12	18	15
S4	1999	20	21	12	15	16	19	18	18	16	9	13	22	17
	2000	19	19	14	14	16	13	17	15	16	10	13	15	15
	2001	16	16	13	13	15	14	19	16	15	11	15	19	15
	2002	20	14	13	10	15	16	15	16	15	14	11	13	14
	2003	21	14	11	11	12	17	13	17	14	11	11	23	15
	2004	18	17	19	16	17	16	16	21	16	17	14	21	17
	2005	20	9	14	11	14	13	16	15	16	11	12	14	14
	2006	16	14	12	10	12	14	16	18	15	14	10	18	14

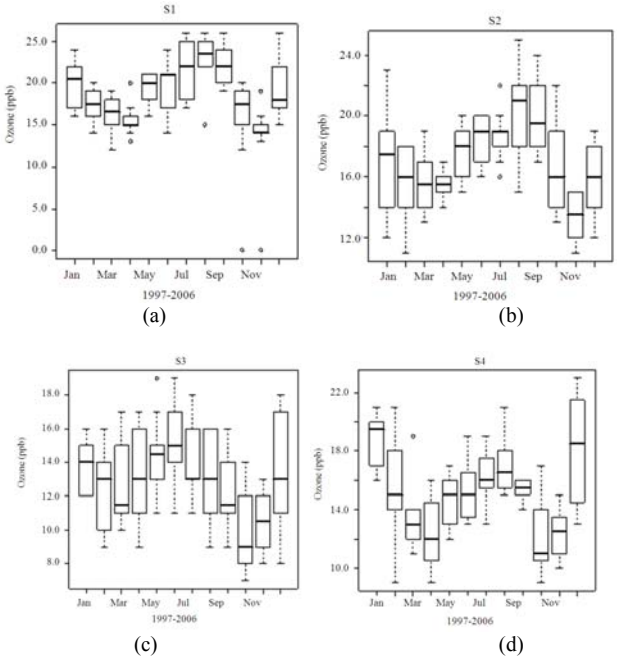

Fig. 3: Box plot of ozone concentrations over an annual cycle during 1997-2006 at selected monitoring stations

Table 3: Correlation (ρ-value) between parameters at monitoring stations

Stations	Parameters	O_3	NO	NO_2	THC
Bukit Kuang (S1)	O_3	1			
	NO	- 0.537**	1		
	NO_2	0.271**	-0.107	1	
	THC	-0.441**	0.342**	-0.359**	1
Indera Mahkota (S2)	O_3	1			
	NO	-0.310**	1		
	NO_2	-0.397**	0.558**	1	
	THC	-0.717**	0.382**	0.411**	1
Kota Bharu (S3)	O_3	1			
	NO	-0.281**	1		
	NO_2	-0.160*	0.589**	1	
	THC	-0.866**	0.109	0.066	1
Kuala Terengganu (S4)	O_3	1			
	NO	-0.579**	1		
	NO_2	-0.272**	0.647**	1	

**: Correlation is significant at the 0.01 level (2-tailed). *: Correlation is significant at the 0.05 level (2-tailed)

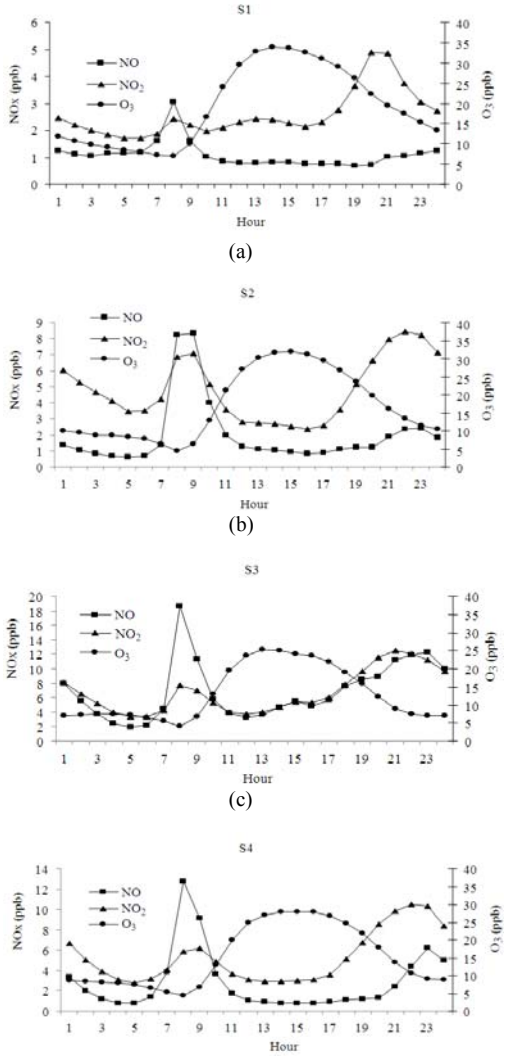

Fig. 4: Diurnal concentration of O_3 compared to NO and NO_2 at selected monitoring stations

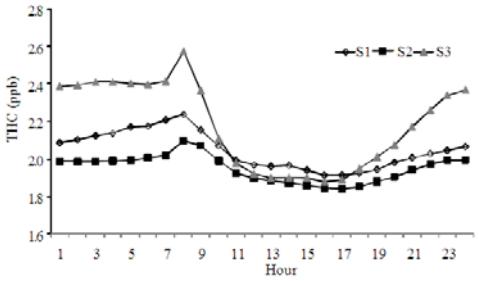

Fig. 5: Diurnal of total hydrocarbon (THC) concentrations recorded at selected monitoring stations (S1, S2 and S3)

value of ρ = -0.579 (negatively correlated) at a significant level of 0.01. Nevertheless, for most of the stations the negative hourly correlation between NO_2 and ozone were not as strong as the correlation between NO and ozone with the exception of Indera Mahkota where the rho value is ρ = -0.310.

The relationship between NO and NO_2 are positively correlated at the highest value of rho,ρ=0.647at Kuala Terengganu (S4) and negatively correlated at the smallest value of rho, ρ = -0.107 at Bukit Kuang (S1).

The ozone concentrations at all stations show a strong negative correlation with Total Hydrocarbon (THC) in the atmosphere. The strongest correlation of ozone and THC was found at Kota Bharu (ρ = -0.866).

Correlation between ozone and meteorological factors (Fig. 6) indicates that the ozone concentration has a significant positive correlation with UV radiation (r = 0.02, p<0.01). Besides, there is a negative correlation between humidity and ozone in ambient air with r = -0.06, p<0.01. The results showed that the cross correlation between ozone and wind speed have a negative correlation with r=-0.109, p<0.01.

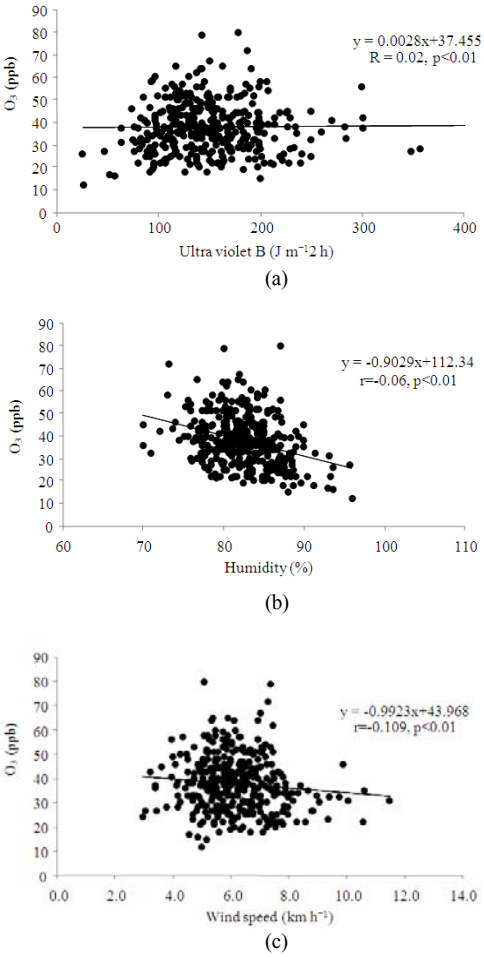

Fig. 6: Correlation between O_3 and meteorolical factors

DISCUSSION

The concentration of ozone recorded at Kota Bharu (S3) and Kuala Terengganu (S4), two stations located in the city centre where there was more traffic, were found to be lower than the values recorded at both Bukit Kuang (S1) and Indera Mahkota (S2). These results concur with research undertaken by Keuken *et al.* (2009), which found that the highest values of ozone are obtained in suburban areas where the influence of road traffic is lower.

The boxplot in the Fig. 3 could indicate the maximum, minimum, mean, 1st and also 3rd quartile values of the concentration. The data that not included between the whiskers is known as outliers with a dot sign.

Strong winds during the monsoon season are capable of transporting ozone the long distances to the monitoring stations as is suggested by Akimoto (1996), Pochanart and Kreasuwun (2001), Lu and Wang (2006), Ishii *et al*. (2007), Al-Jeran and Khan (2009) and Shan *et al*. (2009). These particular seasons (May to September) and (December to March) can also be correlated to biomass burning in Southeast Asia, particularly from Sumatra, Indonesia and Indochina respectively.

High concentrations of NO, are capable of reacting with NO and producing other NO_x, such as NO_2. The amount of ozone found to be reduced by the concentration of NO expected, is contributed to by the exhaust of motor vehicles from the nearby roads as is stated in several other studies (Finlayson-Pits and Pitts 1997; Atkinson, 2000; Al-Azmi *et al*., 2008). Motor vehicles, particularly those moving at low speeds in the city, will emit a high amount of NO into the atmosphere (Ahammed *et al*., 2006). NO than will oxidise to NO_2 in the atmosphere as a result of the reaction with ozone.

The correlation between NO_2 and O_3 recorded at Bukit Kuang was a positive correlation, which indicates that the NO_2 at this station has the ability to produce O_3 in the atmosphere. The concentration of THC recorded at Bukit Kuang (S1), Indera Mahkota (S2) and Kota Bharu (S3) found to have same trends as the concentration of NO (Fig. 5) expected contribute by the number of motor vehicles.

The reaction of NO and O_3 in the atmosphere, as mentioned, will produce NO_2 and this is indicated by the correlation between NO and NO_2. The relationship between NO and NO_2 are positively correlated at Kuala Terengganu (S4) and negatively correlated at Bukit Kuang (S1) demonstrates that the amount of NO to react with O_3 at the rural site of Bukit Kuang is very limited and let to the high concentration of O_3 recorded at this station. This is shown by other studies of O_3 concentrations at rural sites, as reported by Kelly *et al*. (1984) and Al-Azmi *et al*. (2008).

The ozone concentrations at all stations show a strong negative correlation with Total Hydrocarbon (THC) in the atmosphere. The strongest correlation of ozone and THC was found at Kota Bharu. These results concur with those from a number of other studies focused on the correlation between ozone and total hydrocarbon and black carbon in urban areas, e.g., by Latha and Badarinath (2004) and Al-Jeran and Khan (2009).

In order to study ozone variability in East Coast Region of Malaysian peninsula, it is therefore necessary to know the relationships between ozone and the meteorological parameters. The wind is a meteorological variable that strongly affects the ozone concentration, since it determines the transport and dispersion of both ozone and its precursors. Therefore, the correlation between ozone and wind speed is also been studied here.

CONCLUSION

Over the 10-year period (1997-2006), annual mean ozone ranges rose between 12-20 ppb $year^{-1}$ in the East Coast of the Malaysian peninsula. With regard to the trends of ozone at each site, the highest ozone concentrations in the East Coast of the Malaysian peninsula were recorded at Bukit Kuang (S1) with a 10-year mean value of 19 ppb, while the smallest ozone concentrations were at Kota Bharu city centre (S3) with a 10-year mean value of 13 ppb. During this 10-year study period, baseline ozone levels reached a peak of 27 ppb in July 2003, as was documented at Bukit Kuang (S1). In terms of the concentration of primary air pollutants, NO was expected to influence the amount of ozone, specifically at the stations located near too busy roads, NO then oxidised to NO_2 in the ambient air. Our study showed that overall, most of the O_3 recorded at sampling stations had a negative correlation with NO, NO_2 and THC. The research indicates that the ozone level is also affected (positively or negatively) by meteorological conditions, e.g., solar radiation, humidity and wind speed. The results obtained in this study can be used as a physical basis to predict ozone concentration levels in this area.

This study also shows a very strong seasonal variation in ozone concentrations, with higher concentrations observed during the southwest and northeast monsoon. The concentration of ozone was expected to be influenced by the amount of various chemical species in the atmosphere, e.g., CO, NO, NO_x and OH, as a result of oxidation and photochemical processes. The complex reactions due to biomass burning contribute to the long range of ozone distribution and are expected to influence the amount of ozone in the Southeast Asian region.

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