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Fluctuations in nighttime ground-level ozone concentrations during haze events in Malaysia

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Abstract

This study focused on O_3 variations and the titration effects of NO_x during nighttime at urban, industrial, sub-urban and background sites. Nighttime O_3 concentration variations and the presence of high particles with an aerodynamic diameter of less than 10 μm (PM_{10}) were examined because haze disturbs the photochemical reactions of O_3. Hourly data on O_3, NO_2, NO and PM_{10} concentrations provided by the Air Quality Division of the Department of Environment were divided into two groups of daytime and nighttime and analysed. The maximum O_3 concentrations during daytime were generally observed during noon. At nighttime, the concentration of O_3 decreased, indicating that destruction activities occurred mainly via titration. The retention of O_3 during daytime caused the nighttime O_3 during haze events to be higher than that during normal days. Apparent fluctuations in nighttime O_3 concentrations were observed in the urban site (20 ± 13 ppb) during haze events. The NO_2/NO ratio in the urban site during haze was higher than that on normal days; amongst the sites, the urban one had the highest value (6.6). Results indicated that during haze, the reactions between NO and O_3 were enhanced at nighttime, leading to low nighttime NO concentrations. The low nighttime NO concentrations led to low nighttime NO titration rates, which enabled O_3 to persist in ambient air. Nighttime O_3 was not completely absent due to anthropogenic sources. This condition accelerated NO titration to NO_2, thus promoting O_3 production even during haze.

Keywords Anthropogenic sources · Particulate matter · Photochemical reaction · Titration rates · Urban site

Introduction

O_3 exists as a secondary pollutant in the lower atmosphere, where its formation and destruction highly depend on UV radiation and the intensity of its precursors, such as NOx (Ainsworth et al. 2012; Hassan et al. 2013; Alghamdi et al. 2014). Aside from being a secondary pollutant that requires UV light to complete its photochemical reactions, O_3 is a noxious air pollutant and recognised as the second most significant air pollutant in Malaysia (Rahman 2013). O_3 is toxic to humans and vegetation at the ground level due to its capability to oxidise biological tissues (Brimblecombe 2009; Pugliese et al. 2014).

The transformational characteristics of O_3 during haze are crucial for understanding the role of this air pollutant in such an event. Haze conditions may trigger high O_3 photochemical reactions, which intensify the effects of high particulate events resulting from large increments in ambient particulates and O_3. McNaught and Wilkinson (1997) explained that haze is a phenomenon where the readings of an environmental application programme interface consecutively exceed 100 within a 72-h period or longer. They measured the level of visibility interference in the atmosphere by using the coefficient of haze. Quan et al. (2014) reported that stagnation weather, which is generally characterised by low wind speeds and decreased planetary boundary layer height, is the dominant factor in haze events, followed by high particulate emission. Wildfires or biomass burning from the open burning of agricultural residues and forest fires are the major sources of haze, especially in Southeast Asia (Velasco and Rastan 2015; Ahmed et al. 2016). Haze also originates from anthropogenic sources, which are mainly contributed by growing urbanisation and expanding economic activities (Rahman 2013).

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In Malaysia, the high frequency of large-scale biomass burning adversely affects air quality. Several researchers have reported that the long-range transport of air pollution from Sumatera (Ashfold et al. 2017; Kusumaningtyas and Aldrian 2016; Dominick et al. 2015; Khan et al. 2016; Latif et al. 2012) and the prolonged dry weather in Southeast Asia reduce the air quality in Peninsular Malaysia and other neighbouring countries; for example, Singapore experienced extensive haze in June 2013 (DOE 2013; Rahman 2013). Awang et al. (2018) found that light scattering influences the amount of sunlight that penetrates the Earth and the formation of ground-level ozone. Given that ozone requires precursors for their formation, the frequent occurrence of haze as indicated by elevated air pollution index (API) levels essentially represents the rate of transformation of ozone in the atmosphere (Awang et al. 2018).

Theoretically, the O_3 concentration in daytime is higher than that in nighttime because photochemical reactions could occur with the availability of insolation (Alghamdi et al. 2014; Kumar et al. 2015; Reddy and Pfister 2016). The absence of solar radiation during nighttime decreases the O_3 photochemical reaction rate and O_3 concentrations due to continuous chemical destruction by NO and titration (Ghosh et al. 2013). NO_2 does not photolyse at night; as a result, the chemistry of the NO_x family at night is entirely different from that during daytime. The steady state of O_3 concentration is directly proportional to the NO_2/NO ratio (Clapp and Jenkin 2001; Han et al. 2011). Han et al. (2011) reported that O_3 concentration build-up is high in the presence of small values of $[NO_2]/[NO]$ and reaches stable conditions when the O_3 concentration exceeds 90 ppb. However, during nighttime, NO_2 and NO cease to be O_3 precursors and become O_3 reduction agents (Awang and Ramli 2017). Any NO present at night reacts rapidly with O_3 (Eq. (1)). As a result, almost all NO_x at night is converted to NO_2, which reacts with O_3 to produce a nitrate radical (NO_3) (Eq. (2)) (Brown and Stutz 2012) and subsequently removes nighttime O_3 concentration from ambient air.

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (1)$$

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \quad (2)$$

Nighttime processes considerably influence the next day's photochemistry and refine our understanding of the role of heterogeneous processes (reaction between gas- and particulate-phase constituents) occurring at night (Brown and Stutz 2012). Studies have found that up to 50% of freshly emitted NO_x within a 24-h period in urban areas can be removed through nighttime processes (Stutz et al. 2010). NO_x from anthropogenic sources, especially vehicle exhaust, is broken down during nighttime and form NO_3, which is naturally destroyed at night (IDA 2010). Awang et al. (2015) recorded high nighttime O_3 concentrations in Kemaman, Terengganu (Malaysia), due to inefficient removal mechanisms. Ineffective removal or depletion mechanisms cause high O_3 concentrations to remain in the atmosphere and could exert negative effects. Awang and Ramli (2017) emphasized that NO and NO_2 play essential roles in O_3 elimination. Their results confirmed that high nighttime NO concentrations further enhance the destruction of nighttime O_3 concentrations. Dey et al. (2017) found that the diurnal variation of O_3 during nighttime is higher than that during daytime in an industrial site in Golgapur, India. According to Kulkarni et al. (2013), increased nighttime O_3 concentration is associated with prevailing meteorological conditions and transport processes because the photochemical production of O_3 ceases with the absence of sunlight. This explanation could be valid only when the supply of NO_2 at night is constant.

Studies on nighttime O_3 fluctuation during high particulate events (haze) are scarce in Malaysia. Previous studies have confirmed that aerosol particles affect the surface O_3 concentration in Tianjin (Gu et al. 2010; Han et al. 2011). Recently, Awang et al. (2018) reported that haze disturbs the photochemical reactions of O_3. The present study aims to assess variations in nighttime O_3 concentrations during normal days and haze events in Malaysia.

Material and methods

Sampling sites

This study was conducted in four continuous air quality monitoring stations in Malaysia; these stations represent different categories of urban (Seremban (SB)), industrial (Petaling Jaya (PJ)), sub-urban (Muar (MR)) and background (Jerantut (JT)) sites located in Peninsular Malaysia, as shown in Fig. 1. The occurrence of haze for over 24 h was the primary criterion for selecting the study areas because haze is considered to occur once the API value exceeds 100 for 24 h. Figure 2 shows the differences in the trends of PM_{10} concentrations during normal days and haze events.

Sampling measurements

The data for this study, including hourly O_3, NO_2, NO and PM_{10} concentrations, were obtained from the Air Quality Division of the Department of Environment, Ministry of Natural Resources and Environment of Malaysia. These variables (NO_2, NO and PM_{10} concentrations) were selected because of their relationship with O_3 production (Clapp and Jenkin 2001; Ahamad et al. 2014). The haze events in three consecutive years (2013, 2014 and 2015) were regarded as the baseline of nighttime ground-level ozone variations during normal days and haze events. The occurrence of haze for over 24 h was the main criterion for selecting the study areas. Meanwhile, data for normal days refer to the remaining data

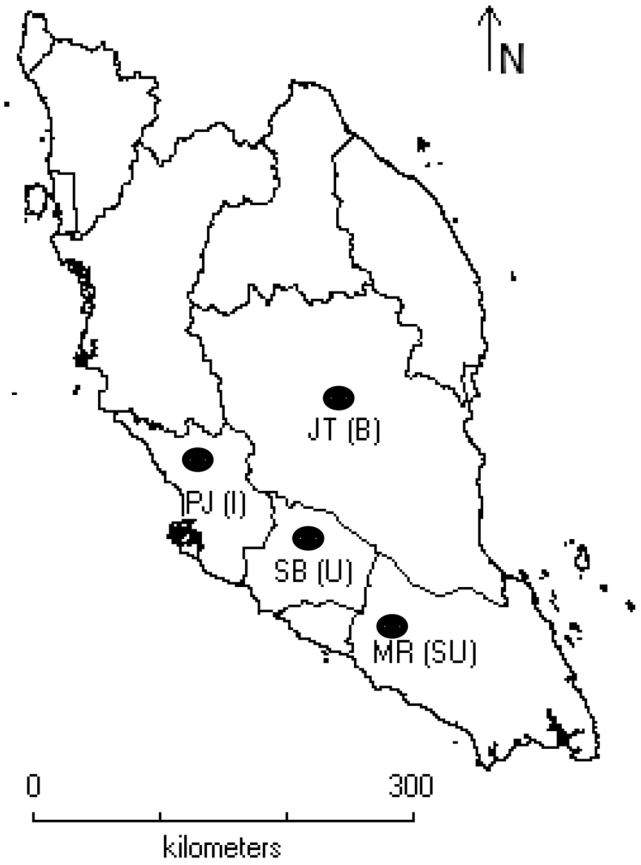

Fig. 1 Location of the study areas at urban, industrial, sub-urban and background sites

for a particular month of the year. Daylight hours (daytime) were defined as the complete hours that fall between sunrise and sunset (Clapp and Jenkin 2001). Nighttime hours ranged from 7 p.m. to 7 a.m. (12 h), as used by Mohammed et al. (2013), Awang et al. (2015) and Awang and Ramli (2017) for the observation of nighttime O_3 diurnal characteristics in Malaysia.

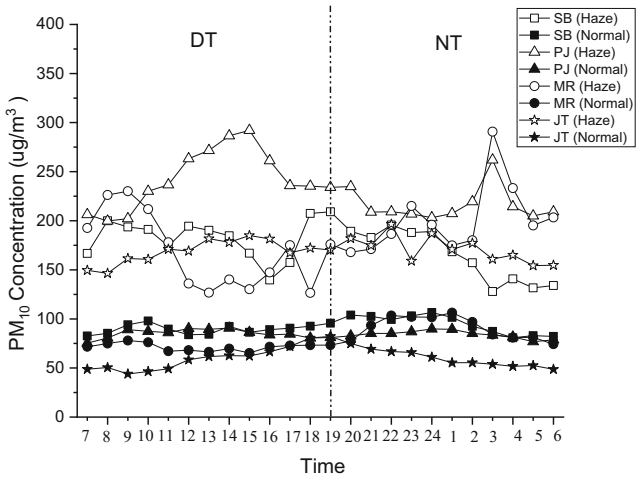

Fig. 2 PM_{10} concentrations at urban, industrial, sub-urban and background sites during normal days and haze events

Hourly O_3 concentration was measured with a UV absorption ozone analyser (Model 400E). The device was operated using the Beer–Lambert Law, which is based on the internal electronic resonance of O_3 molecules with the absorption of 254 nm UV light for measuring low ranges of O_3 concentration in ambient air (Ghazali et al. 2010; Mohammed et al. 2013). NO_2 and NO concentrations were determined using a $NO/NO_2/NOx$ analyser (Model 200A) (Ghazali et al. 2010; Latif et al. 2014). PM_{10} concentration was determined with a BAM-1020 continuous particulate monitor (Met One Instruments, Inc., USA), a high-resolution equipment that can measure PM_{10} concentrations that are as low as 4.8 and 1.0 μg m^{-3} for 1 and 24 h, respectively (Latif et al. 2014).

Results and discussion

Nighttime and daytime ozone measurements

During haze, SB had the highest mean daytime and nighttime O_3 concentrations with a mean value ± standard deviation of 41 ± 16 ppb and 20 ± 13 ppb, respectively, as shown in Table 1. During normal days, given that photochemical reactions occur during daytime, the O_3 concentration was higher than that at nighttime. The nighttime O_3 concentrations observed at this site are higher than the values reported for other cities, such as Kemaman (Awang et al. 2015, 2016), Kolkata (Ghosh et al. 2013) and Portugal (Kulkarni et al. 2013) on a typical day (14, 12 and 10 ppb, respectively). Usually, the lack of human activities at night reduces NO, which eventually leads to low titration (Yusoff et al. 2019).

During haze, the nighttime O_3 concentrations were higher than those on normal days because the efficiency rate of NO_x photochemical reactions increased due to daytime O_3 retention. The variations in nighttime O_3 concentrations in all the sites suggested that the O_3 concentrations during haze were higher than those during normal days. High O_3 concentrations are likely to be found in cities due to high emissions of O_3 precursors (Dufour et al. 2010; Ran et al. 2012) and high occurrence frequency of haze (Awang et al. 2018).

The mean nighttime O_3 concentrations ranged from 10 to 20 ppb. The minimum concentrations were observed from 4 to 6 a.m. Similar to the findings of He et al. (2012), the mean concentrations ranged from 6 to 12 ppb. Saxena and Ghosh (2011) reported that the lifetime of tropospheric O_3 with minor amounts of NO_x is approximately 4–5 days to 1–2 weeks, and hydrocarbons act as an O_3 sink. Meanwhile, at the atmosphere, the lifetime of O_3 is very short (typically hours), especially at high NOx concentrations (Young et al. 2013). These low concentrations of O_3 are due to the absence of photochemical reactions that directly convert O_3 precursors into O_3 (Awang et al. 2015). In the current study, JT (background site) showed the lowest concentrations during normal days

Table 1 Variations in daytime and nighttime O_3 (mean ± standard deviation)

Station (category)	Condition	Daytime O_3 concentration (ppb)	Nighttime O_3 concentration (ppb)
SB (urban)	Normal days	32 ± 14	12 ± 14
	Haze	41 ± 16	20 ± 13
PJ (industrial)	Normal days	31 ± 11	15 ± 6
	Haze	36 ± 12	17 ± 9
MR(sub-urban)	Normal days	26 ± 7	13 ± 7
	Haze	34 ± 12	15 ± 8
JT (background)	Normal days	23 ± 11	10 ± 7
	Haze	36 ± 13	12 ± 7

and haze, with mean nighttime O_3 concentrations of 10 ± 7 ppb and 12 ± 7 ppb, respectively.

Diurnal variation in O_3, NO, NO_2 and PM_{10}

Sunlight induces O_3 photochemical reactions at daytime. Hence, the maximum O_3 concentrations are recorded at noontime in all the sites, as shown in Fig. 3. Meanwhile, during nighttime, O_3 concentrations are theoretically supposed to be zero due to the absence of photochemistry reactions and other destruction processes. However, in this study, high nighttime O_3 concentrations were observed early at night (7 p.m.) and began to decrease until 10 p.m., suggesting that destruction activities occurred mainly through titration. Particles influenced the interaction of O_3 and solar radiation, similar to the results of previous modelling experiments (Li et al. 2011; Péré

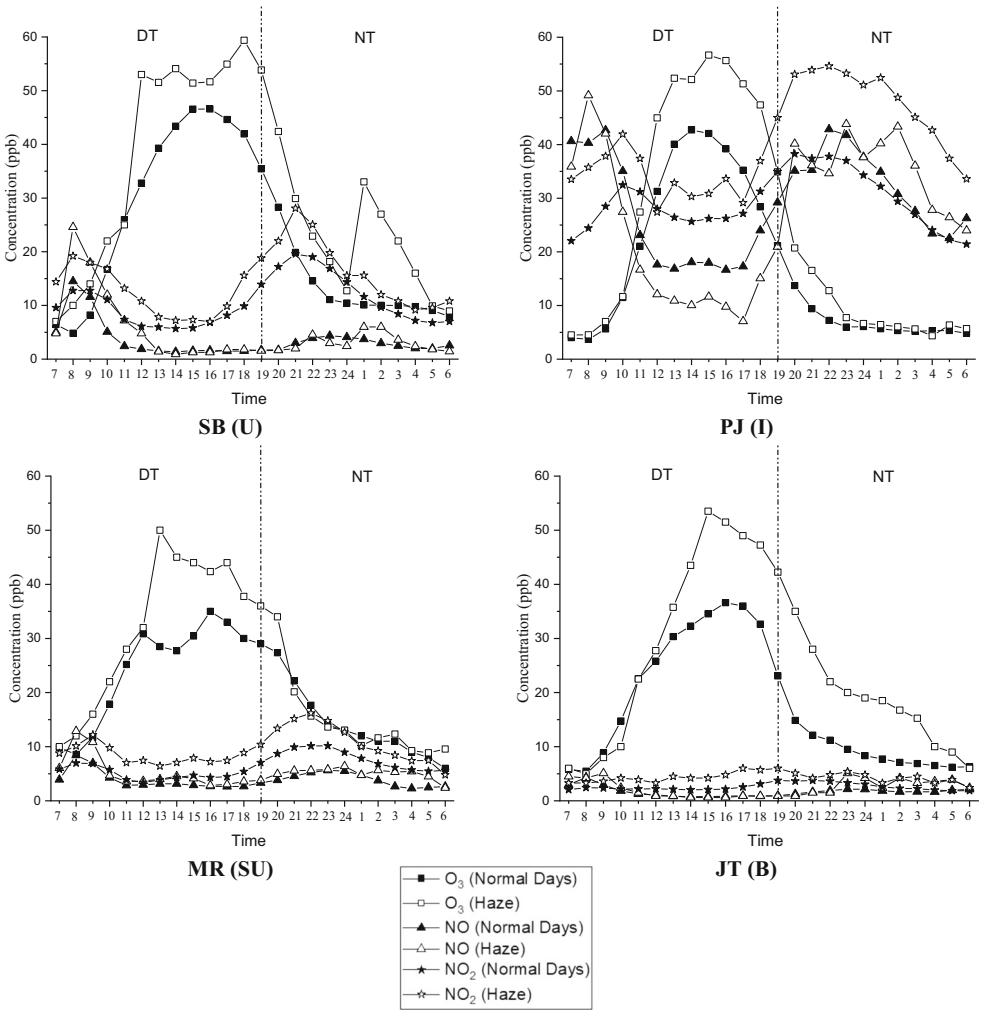

Fig. 3 Diurnal variations of O_3, NO and NO_2 at daytime (DT) and nighttime (NT)

Table 2 Pearson correlation of nighttime O_3 with its precursors and meteorological parameters

Site	Parameter	Normal days						Haze					
		NO	NO_2	PM_{10}	T	RH	WS	NO	NO_2	PM_{10}	T	RH	WS
SB	O_3 (ppb)	**−0.81**	**−0.77**	0.30	**0.94**	**−0.94**	**0.94**	**−0.81**	**−0.64**	**0.52**	**0.93**	**−0.95**	**0.85**
(U)	NO (ppb)	1	**0.72**	−0.16	**−0.75**	**0.76**	**−0.77**	1	0.48	−0.11	**−0.75**	**0.76**	**−0.68**
	NO_2 (ppb)		1	0.34	−0.48	0.48	−0.53		1	−0.12	−0.55	0.49	−0.62
	PM_{10} (μg/m^3)			1	0.48	−0.47	0.13			1	0.42	−0.45	0.07
	T (°C)				1	**−0.98**	**0.93**				1	**−0.98**	**0.85**
	RH (%)					1	**−0.92**					1	**−0.82**
	WS (m/s)						1						1
PJ	O_3 (ppb)	**−0.90**	−0.08	0.51	**0.93**	**−0.89**	**0.93**	**−0.89**	**−0.68**	**0.69**	**0.86**	**−0.87**	**0.83**
(I)	NO (ppb)	1	−0.10	−0.60	**−0.92**	**0.91**	**−0.84**	1	0.60	**−0.70**	**−0.88**	**0.91**	**−0.63**
	NO_2 (ppb)		1	−0.43	−0.53	0.51	−0.45		1	−0.52	−0.56	0.47	−0.47
	PM_{10} (μg/m^3)			1	**0.80**	**−0.83**	0.51			1	**0.90**	**−0.72**	0.38
	T (°C)				1	**−0.98**	**0.77**				1	**−0.94**	0.60
	RH (%)					1	**−0.88**					1	**−0.67**
	WS (m/s)						1						1
MR	O_3 (ppb)	**−0.72**	−0.65	0.11	**0.90**	**−0.96**	**0.94**	**−0.69**	**−0.68**	**0.77**	**0.91**	**−0.95**	**0.90**
(SU)	NO (ppb)	1	**0.83**	−0.07	**−0.81**	**0.79**	**−0.75**	1	**0.78**	0.57	**−0.76**	**0.77**	**−0.78**
	NO_2 (ppb)		1	0.07	**−0.76**	**0.82**	**−0.75**		1	0.65	**−0.73**	**0.76**	**−0.71**
	PM_{10} (μg/m^3)			1	**−0.68**	**0.81**	**−0.70**			1	**−0.68**	**0.75**	−0.61
	T (°C)				1	**−0.96**	**0.99**				1	**−0.98**	**0.99**
	RH (%)					1	**−0.97**					1	**−0.99**
	WS (m/s)						1						1
JT	O_3 (ppb)	**−0.85**	0.08	**−0.81**	**0.99**	**−0.98**	**0.80**	**−0.88**	**−0.69**	**0.86**	**0.99**	**−0.97**	**0.62**
(B)	NO (ppb)	1	−0.03	**−0.73**	**−0.92**	**0.85**	**−0.90**	1	−0.58	**−0.84**	**−0.91**	**0.80**	**−0.82**
	NO_2 (ppb)		1	**0.82**	**0.72**	**−0.74**	0.41		1	0.45	**0.71**	**−0.74**	0.21
	PM_{10} (μg/m^3)			1	**0.87**	**−0.80**	**0.90**			1	**0.88**	**−0.78**	**0.79**
	T (°C)				1	**−0.98**	**0.84**				1	**−0.96**	**0.66**
	RH (%)					1	**−0.67**					1	−0.46
	WS (m/s)						1						1

*Bold with significant value at $p < 0.05$

et al. 2015) that showed that particles decrease daytime O_3 concentrations by 2–17%. The O_3 nighttime chemistry is a new piece of the air quality puzzle. According to Brown and Stutz (2012), nighttime cleansing is slowed down by 7% by the effects of light pollution in Los Angeles, thus increasing O_3 formation the next day by up to 5%. Similarly, nighttime O_3 variation was observed in the current study during haze, and the concentration recorded during haze was higher than that during normal days at all sites. The nighttime O_3 concentrations during haze were influenced by the rate of removal reactions but were unchanged even under high NO_x reactions.

During daytime, the NO_2 peaks occurred about 1–2 h after the NO peak, and the O_3 peak emerged 6 h after the NO peak (Han et al. 2011; Banan et al. 2013). At nighttime, the NO_2 peaks usually occurred 3 h before the NO peak and about 2 h after the O_3 peak in all the sites. This pattern may be due to the nocturnal planetary boundary layer, where the surface emissions of NO are retained under this inversion and cause the hourly NO_x concentration to increase during nighttime. In many instances, these variations depend on site location, local emission of precursor gases (An et al. 2015; Wang et al. 2017; Zhao et al. 2018) and meteorological conditions (Yusoff et al. 2019; Reddy and Pfister 2016; Li et al. 2017).

NO and NO_2 during nighttime in the current study exhibited the same trend as those in the work of Han et al. (2011), Awang and Ramli (2018) and Awang et al. (2018). They began increasing with the destruction of O_3. The variations during normal days and haze were similar, and NO_2 was always higher than NO. Two peaks (i.e. 8–9 a.m. and 9–10 p.m.) were observed in all the sites, and the maximum NO_2 mean concentrations ranged from 5 to 47 ppb. The fluctuation in the urban site was clearer than that in the industrial,

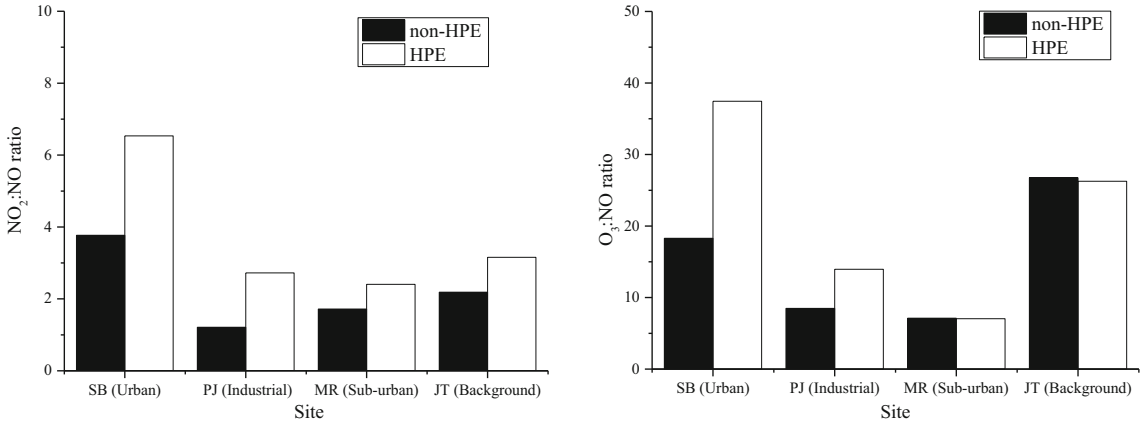

Fig. 4 Ratio of NO_2/NO and O_3/NO

sub-urban and background sites during haze. This finding is similar to that obtained by Awang et al. (2018), who reported that urban sites have higher PM_{10} concentrations than other sites.

Table 2 shows the Pearson correlation results of nighttime O_3 concentrations with NO, NO_2, PM_{10}, temperature, relative humidity and wind speed. During nighttime, temperature exerted a direct effect on O_3 formation (positively correlated). During haze, O_3 and PM_{10} were positively correlated. The highest association was recorded in the background site (JT with 0.86), and the urban site (SB) exhibited the lowest association of 0.52. Meanwhile, a negative correlation between O_3 concentration and NO, NO_2 and relative humidity was found during normal days and haze in all the sites. Thus, the production of O_3 during nighttime could be associated with the decrease in NO, NO_2 and relative humidity. High relative humidity could enhance O_3 cleansing through wet deposition. Similar findings were obtained by Toh et al. (2013) and Awang et al. (2015).

Nighttime NO_2/NO and O_3/NO ratio

Figure 4 shows the mean ratios of NO_2/NO and O_3/NO for all the sites. The concentrations of O_3 increased as the ratio of NO_2/NO increased. All sites showed higher NO_2/NO ratios during haze than during normal days, and the urban site (SB) had the highest value (6.6). These results suggest that haze enhanced the reactions between NO and O_3, resulting in reduced nighttime NO in air. Atkinson (2000) indicated that during nighttime, NO reacts with O_3 and produces NO_2, which in turn produces NO_3 and N_2O_5. Evidently, net production of O_3 still occurs after these conversions.

In the present study, a high NO_2 was recorded in the urban site (SB) due to the contribution of anthropogenic sources, particularly from the downtown site, during haze. Given that NO titration is limited during haze (Awang et al. 2018), the conversion of NO into NO_2 is also limited. This observation is supported by the findings of Khalil et al. (2018). They studied nighttime O_3 concentrations in Makkah and found that nighttime activities in all cities cause high NO_2 and NO concentrations and contribute to the high O_3 concentration at night. Low nighttime NO titration rates could reduce nighttime O_3 (Banan et al. 2013), thus enabling O_3 to persist in ambient air.

Conclusions

This study revealed fluctuations in nighttime O_3 concentrations, especially in urban sites (SB). The mean nighttime O_3 concentrations ranged from 10 ± 7 ppb to 20 ± 13 ppb. The minimum levels during daytime were observed from 4 to 6 a.m., with the mean levels ranging from 6 to 12 ppb. The daytime O_3 level affected the nighttime O_3 level even during haze but at different magnitudes; the value during haze was higher than that during normal days in all the sites. All of the sites showed higher NO_2/NO ratio during haze than during normal days, with the urban site (SB) having the highest recorded value (6.6). Increased NO may result in high levels of nighttime O_3. During haze, the enhanced reactions between NO and O_3 at nighttime lead to low nighttime NO concentrations. Thus, we suspect that additional anthropogenic sources accelerate NO titration to NO_2 and directly increase the O_3 concentrations during nighttime in haze events.

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