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Drivers of nocturnal interactions between ground-level ozone and nitrogen dioxide

Shith S.[1], Ramli N.A.[1], Nadzir A.U.M.[2], Awang N.R.[2] and Ismail M.R.[3*]
[1]Environmental Assessment and Clean Air Research, School of Civil Engineering, Engineering Campus, Universiti Sains Malaysia, 14300, Nibong Tebal, Penang, Malaysia
[2]Faculty of Earth Science, Universiti Malaysia Kelantan Kampus Jeli, 17600, Jeli, Kelantan, Malaysia
[3]School of Housing Building and Planning, Universiti Sains Malaysia, Penang 11800, Malaysia
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*to whom all correspondence should be addressed: e-mail: rodzi@usm.my
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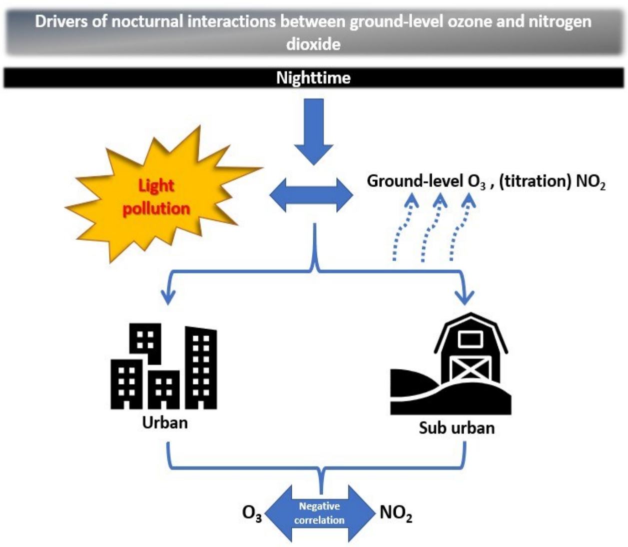
Graphical abstract



Abstract

This study analyses nighttime (7 p.m. to 6 a.m.) light pollution effects on ground-level ozone production in urban and sub-urban sites in Malaysia. In the absence of solar radiation, no photochemical reaction will occur during nighttime, resulting in zero readings of ground-level ozone (O_3). O_3 and nitrogen dioxide (NO_2) data collected in 2020 were analysed to assess time series and diurnal variability at each site and between sites. The highest nighttime mean O_3 concentration is Minden (60 ppb); meanwhile, for nighttime mean NO_2 concentrations, the highest is Klang (43 ppb). The results show that sub-urban experienced higher O_3 variations compared to urban areas. The monthly mean nighttime O_3 and NO_2 in urban and suburban areas display a gradual increase in O_3 and NO_2 variations from March to April, followed by a decreasing trend in the mid of the year. These variations in monthly air pollutants are related to the MCO, CMCO, and RMCO in Malaysia during 2020. Putrajaya (sub-urban site) was the darkest site (average lux: 20) for the whole dataset. In contrast, Minden (sub-urban site) was recorded as the brightest site with maximum light pollution (average lux: 70). The relationship between O_3 and NO_2 shows a negative correlation during nighttime for urban and suburban sites. Light pollution can reach levels that might affect nocturnal O_3 and NO_2 concentrations; therefore, the long-term variability of light pollution is essential for air pollution studies.

Keywords: Anthropogenic source, light intensity, light pollution measurement, nighttime ozone, troposphere ozone

1. Introduction

At the ground level, photochemical reactions of nitrogen oxides (NOx) and volatile organic compounds (VOC) are essential factors contributing to O_3 production in the presence of sunlight (Qiu *et al.*, 2019). Ground-level O_3 has been found to give harmful effects on animals and plants (Unger *et al.*, 2020; Zhang *et al.*, 2021) and caused climate change (Schnell, 2016). In populated and economic zones such as urban and industrial areas, high O_3 pollution occurred due to the emission of NOx and VOCs (Awang *et al.*, 2015; Wang *et al.*, 2019) at downwind locations due to the transport of precursors (Agudelo-Castaneda *et al.*, 2014), meteorological conditions with high temperatures and solar radiation intensities (Han *et al.*, 2011; Castell-Balaguer *et al.*, 2012).

Recently, due to the high nighttime O_3 concentration phenomenon in many regions, focused nighttime O_3 pollution has been crucially observed and investigated (Sousa *et al.*, 2011; Awang *et al.*, 2015; Awang & Ramli, 2017; Shith & Ramli, 2019; Shith *et al.*, 2021; Shith *et al.*, 2022; Wang *et al.*, 2022). Sousa *et al.* (2011) found that meteorological factors increased nighttime O_3 concentrations in urban and suburban areas. With the absence of sunlight during nighttime, the high nighttime concentrations are associated with the transport process and meteorological conditions (Kulkarni *et al.*, 2013). Substantial research also argues that the possibility of light pollution in cities enhanced the O_3 formation during nighttime (Stark *et al*., 2010; Brown *et al*., 2012; Shith *et al*., 2022).

Light pollution, resulting from the alteration of natural night light levels by artificial light sources, is one of the most important pollutants in the atmosphere (Masseti, 2020; Czarnecka *et al*., 2021); it is continuously increasing due to the rising efficiency in producing light (ALAN; artificial light at night). This alteration is originated from the irradiance and glare of lamps. Also, it comes indirectly from the scattering of light in the atmosphere (skyglow), thus affecting the night sky and the biodiversity of rural and natural areas. Night sky brightness positively correlates with several atmospheric parameters, particularly aerosol optical depth and particulate matter (Posch *et al*., 2018; Kocifaj & Barentine, 2021).

Artificial light directly degrades the natural moonlight in the environment, leaving a 'window' for ground-level ozone to be produced even at nighttime. Characterising the level and variability of light pollution has become an important issue for several disciplines, including enhancing the O_3 formation at night (Stark *et al*., 2010; Massetti, 2020). This study aims to investigate the annual/seasonal variability and model the possible relationship between nighttime light pollution and ground-level O_3 and NO_2 variations in urban and suburban areas in 2020, including during Movement Control Order (MCO), Conditional Movement Control Order (CMCO) and Recovery Movement Control Order (RMCO) in Malaysia.

2. Materials and methods

2.1. Urban and sub-urban location

Four (4) sites were chosen to represent the urban and suburban sites. Klang and Shah Alam were selected for the urban site, while Putrajaya and Minden were selected for sub-urban sites. The specific details for each area are depicted in Table 1 with the details on the Bortle dark-sky scale, artificial brightness, artificial lights that increase the night sky luminance and brightness, and the natural brightness of the night sky for both sites. Klang and Shah Alam have high population and traffic density compared to the sub-urban areas of Putrajaya and Minden. Rapid development has accelerated the industrial activities such as manufacturing, factories, processing, shipping, and tourism (Othman & Latif, 2020). The urban area was facilitated by more artificial light sources; streetlights, security lights, lights on vehicles and lighted buildings that may contribute to light pollution that varies in many degrees (Faid *et al*., 2016).

Table 1. Description of the location and brightness of the selected sampling sites

Group	Station	Coordinate	*Bortle Dark-Sky Class	**Artificial Brightness (µcd/m²)	**Brightness (µcd/m²)
Urban	Klang	N03°00.620 E101°24.484	8-9(City sky)	5320	5490
	Shah Alam	N03°04.636 E101°30.673	8-9(City sky)	5930	6100
Sub-urban	Putrajaya	N02°54'55.5 E101°41'25.8	8-9(City sky)	6350	6520
	Minden	N05°21.528 E100°17.864	7 (sub-urban)	2990	3170

**Referring to Bortle (2001); ** referring to light pollution map (Stare, 2021)*

Table 2. Duration of MCO, CMCO and RMCO in 2020 (Mohd Nadzir *et al*., 2021)

Phase	Date	Day
Movement Control Order (MCO)		
Phase 1	18 March 2020–31 March 2020	14
Phase 2	1 April 2020–14 April 2020	14
Phase 3	15 April 2020–28 April 2020	14
Phase 4	29 April 2020–3 May 2020	5
Conditional Movement Control Order (CMCO)		
Phase 1	4 May 2020–12 May 2020	14
Phase 2	13 May 2020–9 June 2020	28
Recovery Movement Control Order (RMCO)		
Phase 1	10 June 2020–31 August 2020	82
Phase 2	1 September 2020–31 December 2020	122
Phase 3	1 January 2021–31 March 2021	90
Movement Control Order (MCO2) by states		
Each state switched between MCO, CMCO, RMCO	11 January 2021–31 May 2021	141
	Movement Control Order (3)	
MCO 3	1 June 2021–28 June 2021	28
	Enhanced Movement Control Order (EMCO)	
EMCO	3 July 2021 onwards	-

2.2. O_3 and NO_2 data collection

The secondary data from 1st January 2020 to 31st December 2020 were used in this study and obtained from the Department of Environment, Malaysia (DoE, 2020). This data was collected during the COVID-19 outbreak under Movement Control Order (MCO), Conditional Movement Control Order (CMCO) and Recovery Movement Control Order (RMCO) in Malaysia, as shown in Table 2. The data were grouped as nighttime hourly average (7 p.m. to 6 a.m.) for the analysis. The hourly average of O_3 and NO_2 was measured using a UV photometric Thermo Scientific Ozone Analyzer (Model 49i). NO concentration was unavailable for 2020, thus the results depended on the recorded O_3 and NO_2 concentrations.

2.3. Light intensity data collection

The lux reading for light pollution was carried out using the portable lux meter HI97500. The instrument is supplied with a light sensor connected by a fixed 1.5 m coaxial cable to allow measurements to be taken from a distance without user interference. By simply pressing the RANGE key, users can switch among three ranges to choose the best resolution according to the tested environment. The HI97500 lux meter has a rugged and water-resistant body for frequent outdoor use. The HI97500 features a low battery indicator and automatic shut-off that turns the meter off after 7 minutes of non-use. This method was implemented at a horizontal plane around 0.8 m above the ground level (floor). The light sensor of the instrument was placed on any horizontal plane to avoid obstructing the typical light path (the path between the lighting source and the light sensor should be clear as far as practicable). In this research, the lighting assessment was evaluated during the nighttime on all artificial lights near the air quality stations. Five (5) readings of lighting level with 30 seconds time intervals were used to measure the light reading, as shown in Table 3.

Table 3. Lux reading under the lamp post at urban and sub-urban sites

Area	Site	Time	Lamp Post	1	2	3	4	5	Average (Avg)		Avg
Urban	Klang	1940 - 2020	A	0.048	0.048	0.048	0.048	0.048	0.048	48.00	52.95
			B	0.048	0.049	0.049	0.049	0.049	0.049	48.80	
			C	0.059	0.059	0.059	0.060	0.060	0.059	59.40	
			D	0.055	0.056	0.056	0.056	0.055	0.056	55.60	
	Shah Alam	2045 - 2125	A	0.024	0.024	0.024	0.024	0.024	0.024	24.00	22.50
			B	0.021	0.021	0.021	0.021	0.021	0.021	21.00	
			C	0.025	0.025	0.025	0.025	0.025	0.025	25.00	
			D	0.020	0.020	0.020	0.020	0.020	0.020	20.00	
Sub-urban	Putrajaya	(1845). - 1940	A	0.016	0.016	0.016	0.016	0.017	0.016	16.20	19.43
			B	0.020	0.020	0.020	0.020	0.020	0.020	20.00	
			C	0.024	0.024	0.024	0.023	0.023	0.024	23.60	
			D	0.064	0.064	0.064	0.064	0.063	0.064	63.80	
			E	0.000	0.000	0.000	0.000	0.000	0.000	0.00	
			F	0.009	0.009	0.009	0.010	0.010	0.009	9.40	
			G	0.003	0.003	0.003	0.003	0.003	0.003	3.00	
	Minden		Five lamp post				-				70.00

2.4. Statistical analysis

The box of whisker plots and time series plots were used to visualise the seasonal, annual, monthly and diurnal trends of nighttime ground-level O_3 and NO_2 concentrations. A general linear regression analysis of nighttime ground-level O_3 and NO_2 concentrations was conducted to analyse the significance or persistence between both pollutants. Statistical analysis in this study was conducted with Origin Pro version 10 software.

3. Results and discussion

3.1. Nighttime ground-level O_3 and NO_2 characteristics

Figure 1 shows the box of whisker plot of ground-level O_3 and NO_2 at urban and suburban sites. The highest nighttime mean ground-level O_3 concentrations were recorded in suburban areas compared to urban areas. The highest nighttime mean ground-level O_3 concentration is Minden (60 ppb), followed by Shah Alam (58 ppb), Putrajaya (54 ppb) and Klang (50 ppb). Meanwhile, for nighttime mean NO_2 concentrations, the highest is Klang (43 ppb), followed by Shah Alam (40 ppb), Putrajaya (30 ppb) and Minden (20 ppb). The high O_3 concentrations in urban sites (Shah Alam) happened before the MCO was implemented. This is due to various anthropogenic activities which became a significant source of O_3 precursors in urban areas (Ghazali *et al*., 2010; Latif *et al*., 2012; Awang *et al*., 2015; Shith *et al*., 2021; Shith *et al*., 2022). After the MCO, the O_3 concentrations reduced significantly, indicating the reduction in motor vehicles on the road as the effect of the temporary closure of industries during the MCO 1 (Othman & Latif, 2020; Mohd Nadzir *et al*., 2021; Latif *et al*., 2021; Awang *et al*., 2022).

Figure 1 also shows the monthly/annual variations of ground-level O_3 and NO_2 concentrations. In both urban and sub-urban sites, there are different variations. The monthly mean nighttime ground-level O_3 and NO_2 in urban and suburban areas display a gradual increase in ground-level O_3 and NO_2 variations from March to April, followed by a decreasing trend in the mid of the year. These variations in monthly air pollutants are related to the MCO, CMCO and RMCO in Malaysia during 2020. Conversely, the nighttime NO_2 trend for the sub-urban is more stable compared to the urban sites, as depicted by the box plot. The box showed high variations in Klang, and Shah Alam compared to Putrajaya and Minden. The monthly nighttime variations in NO_x were similar to other studies (Guttikunda & Gurjar, 2011; Awang *et al*., 2015; Shith *et al*., 2022), which attributed to the poor dependence of NO_x on the meteorological situation. Less anthropogenic sources in sub-urban were related to the decrease of NO_2 during nighttime.

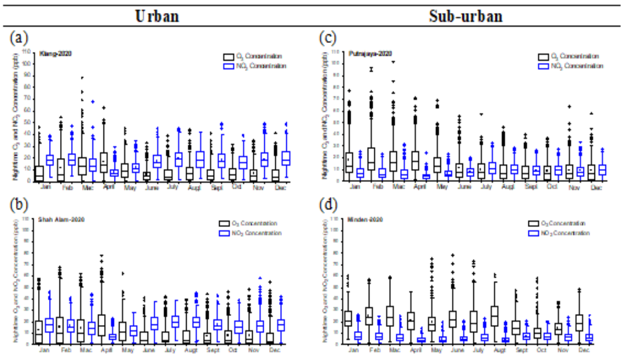


Figure 1. Box of whisker plots of nighttime O_3 and NO_2 for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

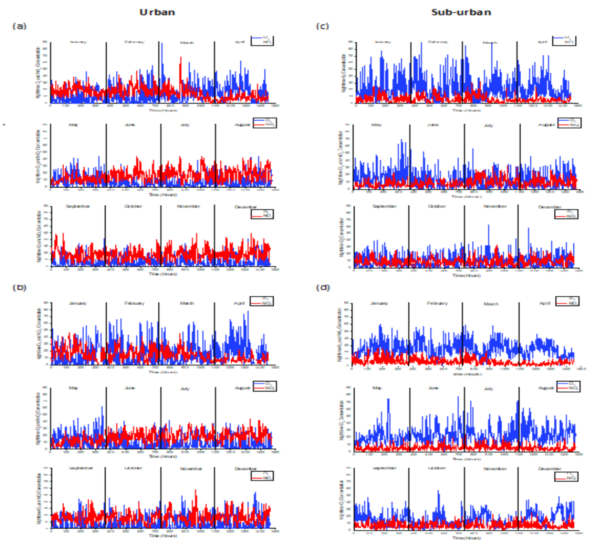


Figure 2. Time Series plot of Nighttime O_3 and NO_2 Concentration for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

3.2. Time series and diurnal variations

The variability of nighttime mean ground-level O_3 concentrations in urban and sub-urban sites was further investigated by plotting the time-series trend using the hourly average nighttime O_3 concentrations, as shown in Figure 2. The figure shows that sub-urban sites have higher nighttime mean ground-level O_3 concentrations than NO_2. This trend differed with urban areas, whereby the nighttime ground-level O_3 and NO_2 concentrations exhibited the same trend. The fluctuation of nighttime O_3 concentrations was observed from January to December 2020. The result showed that O_3 concentrations were significantly higher from January to March, which is the period when the MCO was enacted. Right after the implementation of MCO, O_3 concentrations were consistently low due to the low emission of its precursors.

The diurnal variations of nighttime ground-level O_3, NO_2 and light pollution are presented in Figure 3. Average light pollution was significantly different among the sites. Putrajaya (sub-urban site) was the darkest place (average lux: 20) for the whole dataset. In contrast, Minden (sub-urban site) was recorded as the brightest place with maximum light pollution (average lux: 70). Unexpectedly, high light pollution occurred in Minden (sub-urban area). This could be due to the location itself, as Minden was located in the university area, which was highly illuminated for safety and security reasons of the students during nighttime. Similar observations have been recorded by Shith *et al*. (2022) at Putrajaya, as it functions as the federal administrative centre in Malaysia. This place was busy during the daytime when people went to work and had fewer activities during the nighttime.

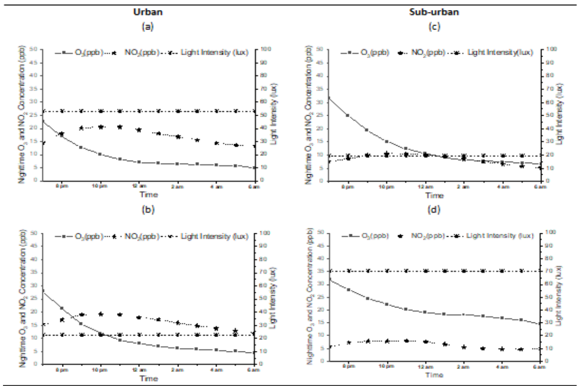


Figure 3. Diurnal variations of nighttime ground-level O_3, NO_2, and light pollution for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

The average urban site light pollution was 25-54 lux lower than in the sub-urban sites. According to a few researchers, weather conditions, masked by clouds and particularly precipitation, affect the reading of light pollution by increasing it in sub-urban areas, resulting in conditions sub-urban brighter than in urban areas (Puschnig *et al*., 2014; Posch & Puschnig, 2018; Massetti, 2020). The nighttime O_3 concentrations trend decreased rapidly from 7 p.m., then gradually until it reached 6 a.m. of the next day. Both sites depicted the same trend during nighttime. This phenomenon may be attributed to the current decrease in the precursors that leads to decreases in ozone photochemical reactions as it approaches nighttime (Guttikunda an&urjar, 2011; Awang *et al*., 2015; Shith *et al*., 2022). The decrements are also governed by the NO titration proses, which according to Awang *et al*. (2015), is the primary removal reaction of O_3 during nighttime. According to Michael *et al*. (2020) irradiance for visible light is between 380 to 780 nm, while the UVB responsible for ozone photochemical reactions is between 280 to 320 nm. Thus, high-intensity light such as stadium spotlight might have greater irradiance thus possibly having enough energy to enable ozone photochemical radiation during nighttime.

The minimum nighttime O_3 concentrations were recorded at six ppb for urban and suburban sites. The minimum nighttime NO_2 concentrations were maintained until the following day's rush hours at urban and suburban with the recorded concentrations of 12 ppb and six ppb, respectively, due to the oxidation of NO to NO_2 by O_3 during nighttime in the absence of radiation (Song *et al*., 2011). Conversely, NO_2 displayed late afternoon peaks at 10 p.m. for both urban and suburban sites, with concentrations of 20 ppb and 12 ppb, respectively. This peak coincided with rush-hour traffic (Latif *et al*., 2021; Awang *et al*., 2022). Even though the effect of light pollution is small, still, this phenomenon has different consequences for O_3 formation (AGU, 2012), where the lighting in urban sites influences NO_3 photolysis as a sink for NO_3 and N_2O_5 at night (Brown *et al*., 2007; Stark *et al*., 2010).

3.3. Nighttime relationship between O_3 and NO_2

The effect of nighttime O_3 and NO_2 was analysed using a general linear model for urban and sub-urban sites depicted in Figure 4. The O_3 concentrations during nighttime are much lower in urban areas compared to suburban locations. Indicating that even photochemical reactions inhibited at night, O_3 concentrations still exist. The relationship between O_3 and NO_2 has been studied in detail by many researchers (Han *et al*., 2011; Awang *et al*., 2015; Shith *et al*., 2022). O_3 precursors (VOCs, NO_2 and CO), nitrogen oxides (NOx), photochemistry and transport are the main factors for O_3 transformation.

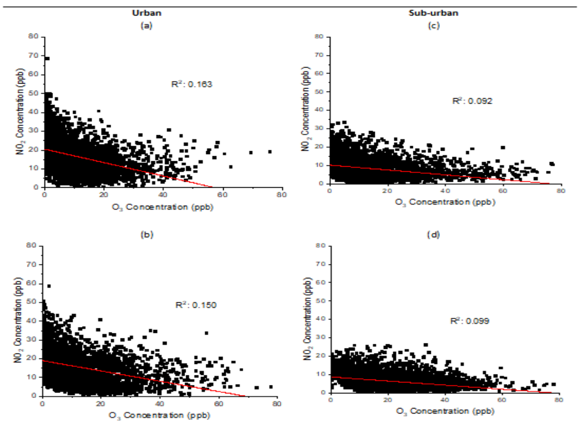


Figure 4. Linear regression of O_3 and NO_2 for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

NO, and NO_2 are known as the main precursors of O_3 concentrations. The figure demonstrated that nighttime O_3 were negatively correlated with NO_2 at urban and suburban sites. The relationship was small with urban sites; Klang and Shah Alam recorded R^2=0.163 and R^2=0.150, respectively. Meanwhile, for sub-urban sites, Putrajaya and Minden recorded R^2=0.092 and R2=0.099, respectively. This is consistent with Abdul Wahab *et al*. (2005) research, which revealed that NO_2 was the primary influence of O_3 formation without solar radiation at night. According to Awang and Ramli (2017), nighttime O_3 chemistry is primarily controlled by the reaction of NO and O_3 concentrations, mainly attributed to the ceasing of photochemical reactions due to the absence of solar radiation. Meanwhile, Shith *et al*. (2022) suggested that minimal O_3 concentrations at nighttime might indicate some light pollution contribution to O_3 formation.

4. Conclusions

Overall, the possible relationship between nighttime light pollution and ground-level O_3 variations between two areas: urban and suburban, in Malaysia has been investigated in this study. The data were grouped as nighttime (7 p.m. to 6 a.m.) to analyse the variations. Remarkably, from the results, sub-urban sites (Minden) had the highest nighttime O_3 because they were located in the middle of the main roads and expressways. Meanwhile, the highest nighttime mean NO_2 concentrations are in urban sites (Klang), at 43 ppb. Thus, weather conditions, masked by clouds and particularly precipitation, affect the reading of light pollution by increasing it in sub-urban areas, making sub-urban conditions brighter than in urban areas. Long-term monitoring and data analysis to characterise the night light intensity is needed to evaluate the light exposure.

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