

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

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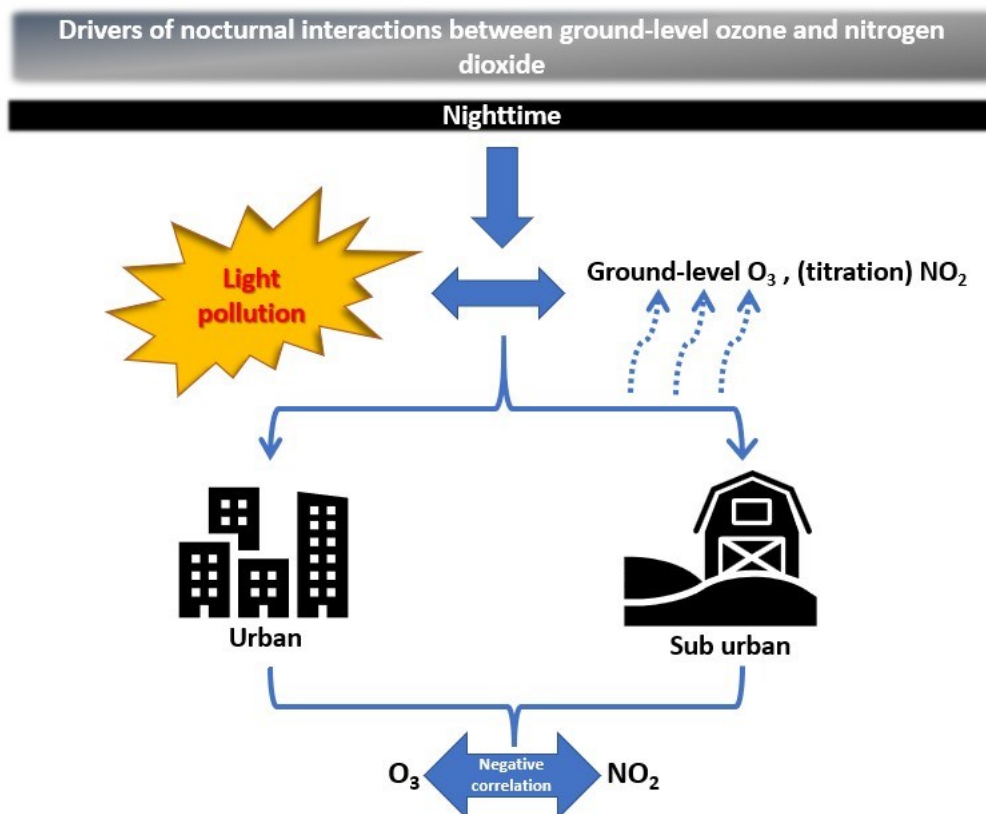
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GRAPHICAL ABSTRACT



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 (NO_x) 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 investigated 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 NO_x 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 O₃ concentration phenomenon in many regions, focused nighttime O₃ 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 are the factors that increased nighttime O₃ 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 O₃ 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 origin 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 O₃ 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 O₃ and NO₂ 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 occurred in industrial activities such as manufacturing, factories, processing, shipping, and tourism (Othman & Latif, 2020). The urban area was facilitated with 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

2.2. O₃ and NO₂ 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 O₃ and NO₂ was measured using a UV photometric Thermo Scientific Ozone Analyzer (Model 49i). NO concentration was unavailable for 2020, so the results depended on the recorded O₃ and NO₂ concentrations.

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

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 nearest 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

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 O₃ and NO₂ concentrations. A general linear regression analysis of nighttime ground-level O₃ and NO₂ 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 O₃ and NO₂ characteristics

Figure 1 shows the box of whisker plot of ground-level O₃ and NO₂ at urban and suburban sites. The highest nighttime mean ground-level O₃ concentrations were recorded in suburban areas compared

to urban areas. The highest nighttime mean ground-level O₃ concentration is Minden (60 ppb), followed by Shah Alam (58 ppb), Putrajaya (54 ppb) and Klang (50 ppb). Meanwhile, for nighttime mean NO₂ concentrations, the highest is Klang (43 ppb), followed by Shah Alam (40 ppb), Putrajaya (30 ppb) and Minden (20 ppb). The high O₃ 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 O₃ 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 O₃ 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 O₃ and NO₂ concentrations. In both urban and sub-urban sites, there are different variations. The monthly mean nighttime ground-level O₃ and NO₂ in urban and suburban areas display a gradual increase in ground-level O₃ and NO₂ 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 NO₂ 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 NO₂ during nighttime.

Figure 1 Box of whisker plots of nighttime O₃ and NO₂ 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 O₃ concentrations in urban and sub-urban sites was further investigated by plotting the time-series trend using the hourly average nighttime O₃

concentrations, as shown in Figure 2. The figure shows that sub-urban sites have higher nighttime mean ground-level O₃ concentrations than NO₂. This trend differed with urban areas, whereby the nighttime ground-level O₃ and NO₂ concentrations exhibited the same trend. The fluctuation of nighttime O₃ concentrations was observed from January to December 2020. The result showed that O₃ concentrations were significantly higher from January to March, which is the period when the MCO was enacted. Right after the implementation of MCO, O₃ concentrations were consistently low due to the low emission of its precursors.

Figure 2. Time Series plot of Nighttime O₃ and NO₂ Concentration for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

The diurnal variations of nighttime ground-level O₃, NO₂ 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, where highly illuminated for safety and security reasons for the students during nighttime. Similar observations have been recorded by Shith et al. (2022) at Putrajaya, as its 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.

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 O₃ 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 and Urjar, 2011; Awang et al., 2015; Shith et al., 2022;). The decrements are also governed by the NO titration process, which according to Awang et al. (2015), is the primary removal reaction of O₃ 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.

Figure 3. Diurnal variations of nighttime ground-level O₃, NO₂, and light pollution for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

The minimum nighttime O₃ concentrations were recorded at six ppb for urban and suburban sites. The minimum nighttime NO₂ 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 NO₂ by O₃ during nighttime in the absence of radiation (Song et al., 2011). Conversely, NO₂ 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 O₃ formation (AGU, 2012), where the lighting in urban sites influences NO₃ photolysis as a sink for NO₃ and N₂O₅ at night (Brown et al., 2007; Stark et al., 2010).

3.3. Nighttime Relationship between O₃ and NO₂

The effect of nighttime O₃ and NO₂ was analysed using a general linear model for urban and suburban sites depicted in Fig. 4. The O₃ concentrations during nighttime are much lower in urban areas compared to suburban locations. Indicating that even photochemical reactions inhibited at night, O₃

concentrations still exist. The relationship between O₃ and NO₂ has been studied in detail by many researchers (Han et al., 2011; Awang et al., 2015; Shith et al., 2022). O₃ precursors (VOCs, NO₂ and CO), nitrogen oxides (NO_x), photochemistry and transport are the main factors for O₃ transformation. NO, and NO₂ are known as the main precursors of O₃ concentrations. The figure demonstrated that nighttime O₃ were negatively correlated with NO₂ at urban and suburban sites. The relationship was small with urban sites; Klang and Shah Alam recorded R²=0.163 and R²=0.150, respectively. Meanwhile, for sub-urban sites, Putrajaya and Minden recorded R²=0.092 and R²=0.099, respectively. This is consistent with Abdul Wahab et al. (2005) research, which revealed that NO₂ was the primary influence of O₃ formation without solar radiation at night. According to Awang and Ramli (2017), nighttime O₃ chemistry is primarily controlled by the reaction of NO and O₃ concentrations, mainly attributed to the ceasing of photochemical reactions due to the absence of solar radiation. Meanwhile, Shith et al. (2022) suggested that minimal O₃ concentrations at nighttime might indicate some light pollution contribution to O₃ formation.

Figure 4. Linear regression of O₃ and NO₂ for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

4. Conclusions

Overall, the possible relationship between nighttime light pollution and ground-level O₃ 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 O₃ because they were located in the middle of the main roads and expressways. Meanwhile, the highest nighttime mean NO₂ 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.

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

Group	Station	Coordinate	*Bortle Dark-Sky Class	**Artificial Brightness ($\mu\text{cd}/\text{m}^2$)	**Brightness ($\mu\text{cd}/\text{m}^2$)
Urban	Klang	N03°00.620	8-9	5320	5490
		E101°24.484	(City sky)		
	Shah Alam	N03°04.636	8-9	5930	6100
		E101°30.673	(City sky)		
Sub-urban	Putrajaya	N02°54'55.5	8-9	6350	6520
		E101°41'25.8	(City sky)		
	Minden	N05°21.528	7	2990	3170
		E100°17.864	(sub-urban)		

*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	-

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	

Sub-urban	Putrajaya	(1845). - 1940	D	0.020	0.020	0.020	0.020	0.020	0.020	20.00	19.43
			A	0.016	0.016	0.016	0.016	0.017	0.016	16.20	
			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

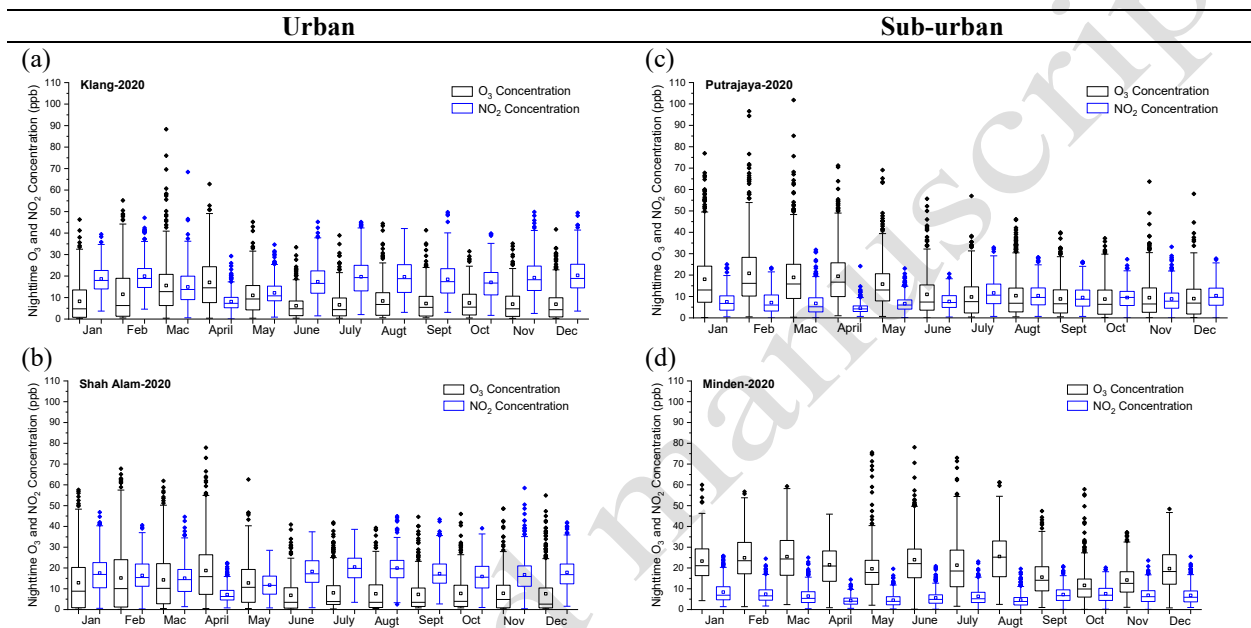
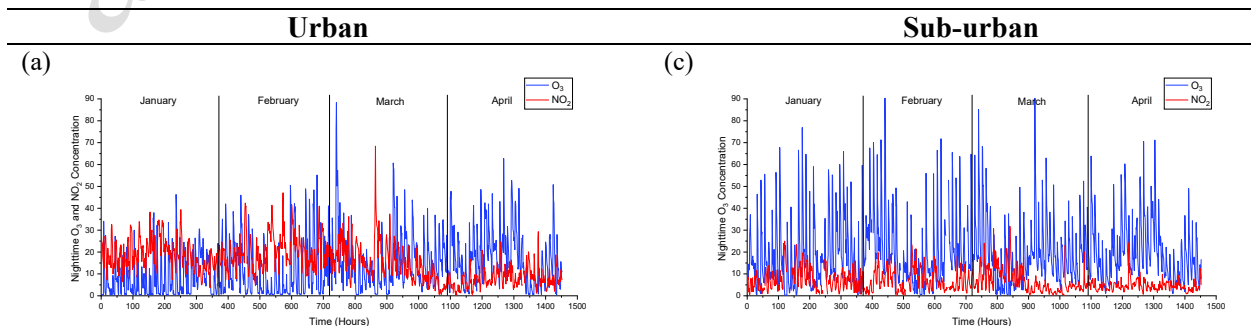


Figure 1. Box of whisker plots of nighttime O₃ and NO₂ for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden



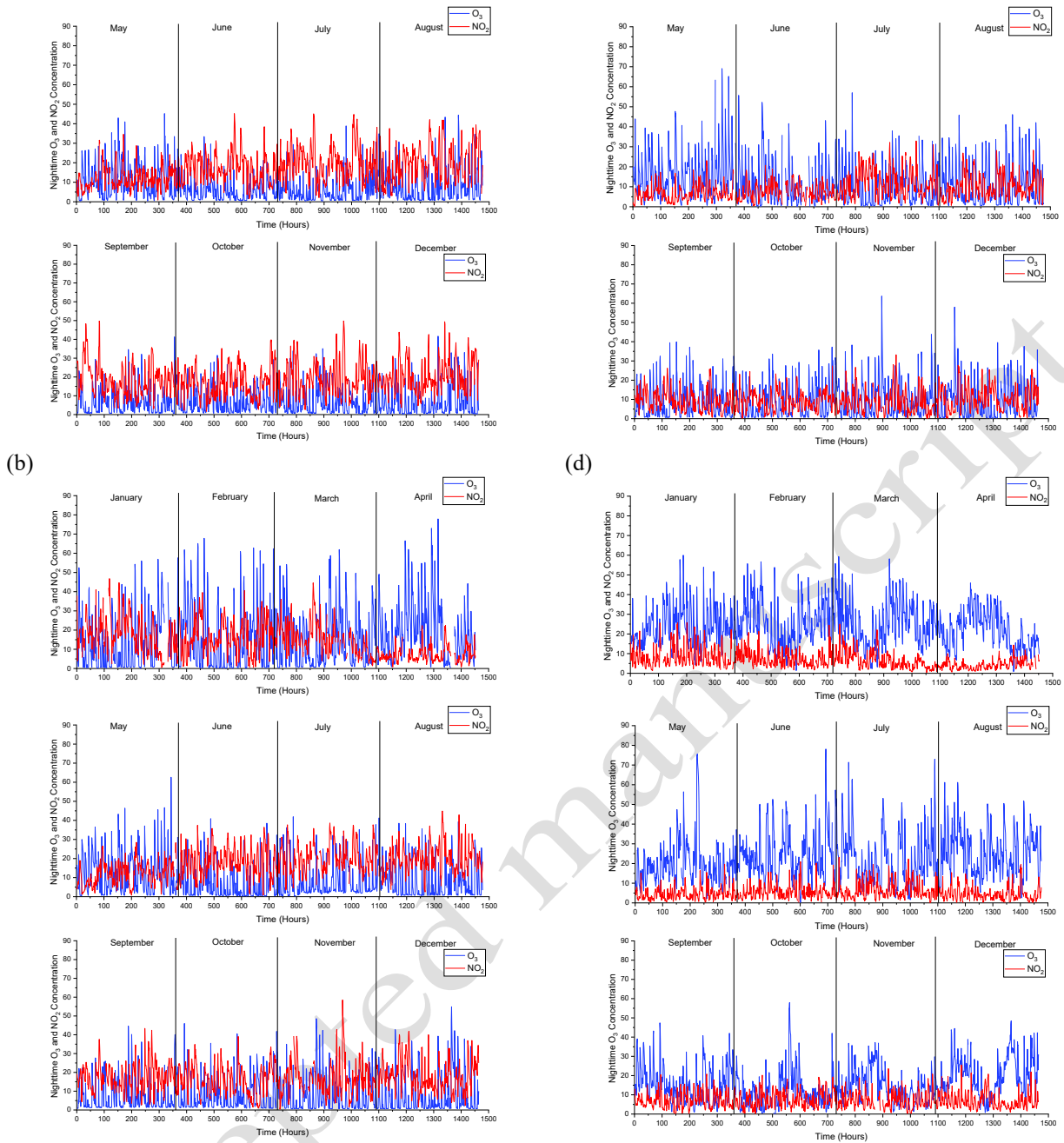


Figure 2. Time Series plot of Nighttime O₃ and NO₂ Concentration for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

Urban

Sub-urban

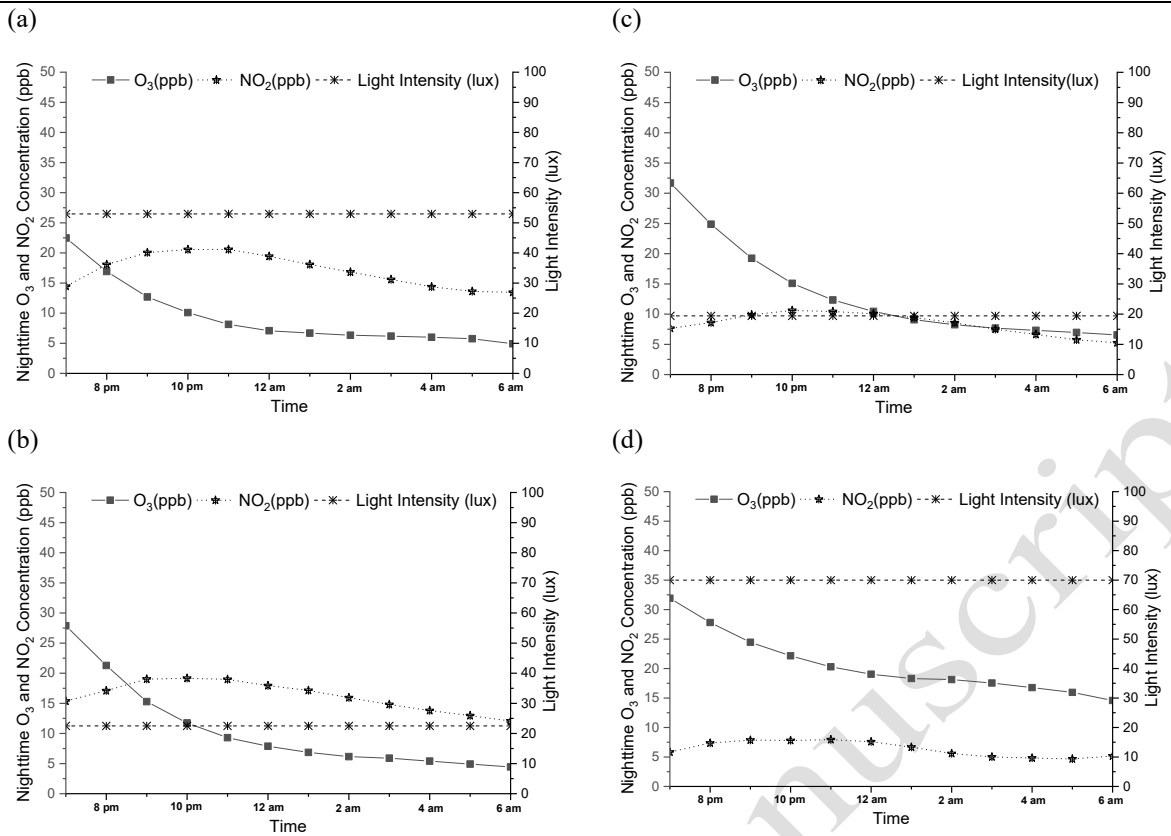


Figure 3. Diurnal variations of nighttime ground-level O₃, NO₂, and light pollution for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

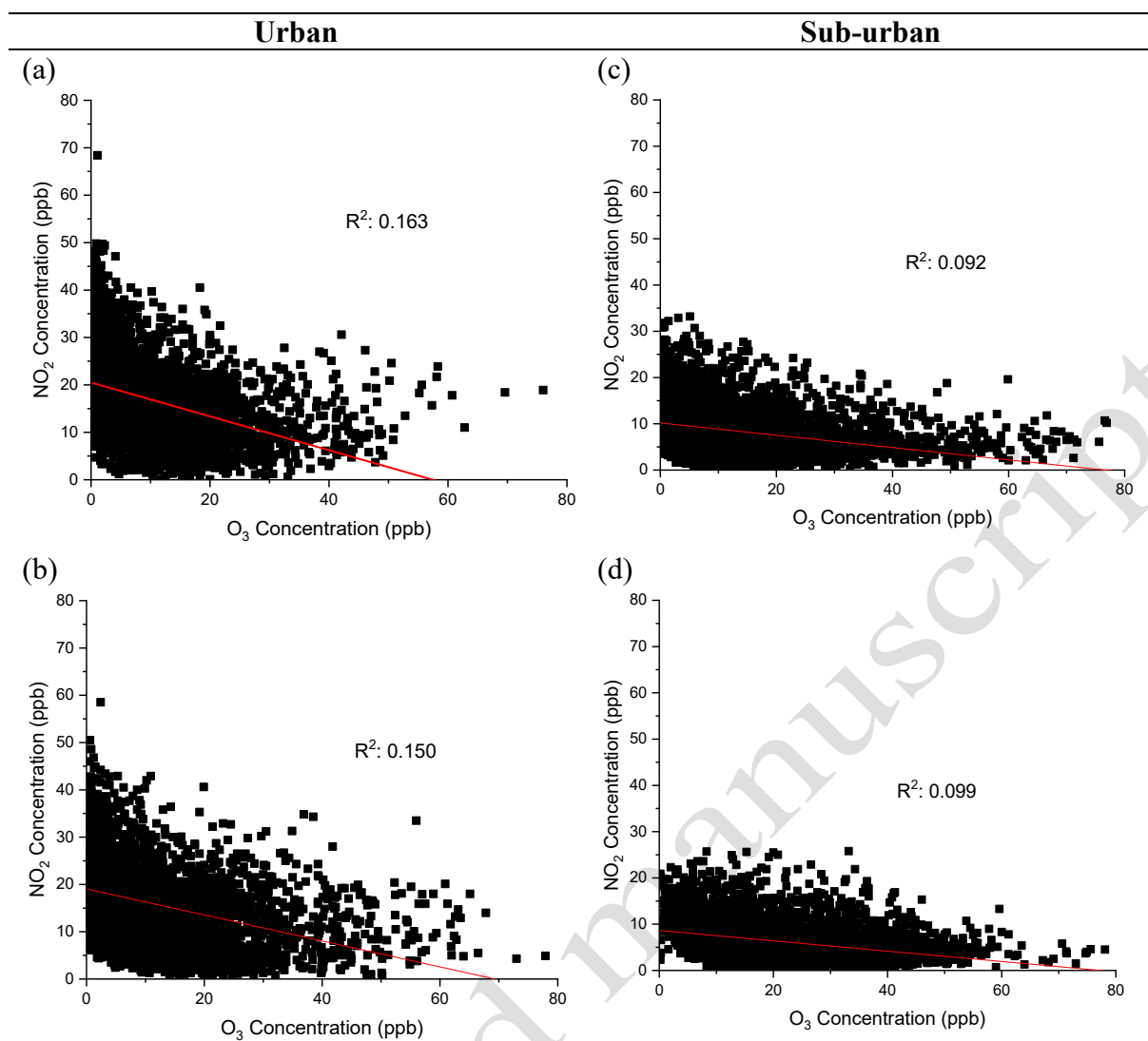


Figure 4. Linear regression of O₃ and NO₂ for 2020 at (a) Klang, (b) Shah Alam, (c) Putrajaya, (d) Minden

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