



Variability of ozone and oxides of nitrogen in the tropical city, Bengaluru, India

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Abstract Bengaluru, also considered India's Silicon Valley, has seen steady growth in population over the years. Bengaluru's rapid development has resulted in dwindling reservoirs, increased traffic congestion, high levels of air pollution, and, to some measure, a rise in summer temperatures. As a result of these changes in urban form over the last decade, anthropogenic heat fluxes for ozone production have increased. However, an observational study on the effects of growing urbanisation on trace gases in Bengaluru for various seasons and periods of the day is missing. Hence, in situ measurements of O_3 , NO, NO_2 , and NO_x concentrations were carried out at Bengaluru, India, from January 2015 to December 2018. The data were examined for diurnal and interannual variations in trace gas mixing concentrations. The diurnal trend in O_3 exhibits unimodal behaviour. Changes in

photochemistry, local meteorology, and the planetary boundary layer's distinctive features cause a rise in the value of concentrations and lead to a peak. In contrast, the diurnal trend in NO, NO_2 , and NO_x displayed a bimodal peak due to the combined effect of vehicular emissions and the planetary boundary layer. The link involving the oxidant OX ($O_3 + NO_2$) and NO_x levels were investigated to determine the NO_x -independent regional and NO_x -dependent local contributions to OX in the atmosphere. Daytime contributions are higher than night-time contributions, according to the present study. The observed anomalies could be the consequence of photochemical processes that produce OX.

Keywords Ozone · Oxides of nitrogen · Trace gases · Oxidant

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Introduction

Ozone (O_3) is a greenhouse gas that is a precursor to the extraordinarily active hydroxyl radical (OH), which controls the chemical characteristics of the troposphere (Pancholi et al., 2018). Variability in the concentration of O_3 is determined by a set of local and global atmospheric processes, including photolysis, its precursor's emissions with vehicular numbers, and density owing to land use land cover (LULC) changes (Chate et al., 2014). Several atmospheric chemical processes control the concentration of O_3 in

the atmosphere. The stratospheric ozone transported downward and in situ formation via the photochemical interaction of carbon monoxide (CO), methane (CH₄), and volatile organic compounds (VOCs) in the presence of Nitrogen Oxides (NO_x) are the two main drivers of tropospheric O₃ levels. Dry deposition or photochemical titration both contribute to the loss of tropospheric O₃ (Varotsos et al., 2000). Due to high solar flux, high humidity, and nitric oxide (NO) and nitrogen dioxide (NO₂) in the atmosphere, the photochemical generation of O₃ is crucial. Plants and biological organisms are harmed by higher O₃ levels in the boundary layer. The hostile effects of surface O₃ levels comprise impairment to plant tissues, natural resources, human's lungs, respiratory problems, and hospitalisations (Tyagi et al., 2020). Because of its ability to induce respiratory disease in humans, ground-level O₃ is also considered a significant air contaminant (Huang et al., 2012).

The nitrogen oxides NO_x (NO_x=NO+NO₂) play a significant role in the oxidising ability of the Earth's lower atmosphere by regulating the sectionalisation, production, and loss of free radical species. NO_x's lifetime varies depending on the photochemical system. It is usually hours to days before it oxidises into HNO₃ and peroxyacetyl nitrate (PAN) (Seinfeld & Pandis, 2006). NO_x has a substantial part in the chemistry of tropospheric O₃ and secondary aerosol production. Sources of NO_x are both natural and anthropogenic. Soil microbiological processes and, to a lesser extent, lightning activity are natural sources, but fossil fuel combustion and biomass burning are manmade activities (Seinfeld & Pandis, 2006). The principal atmospheric NO_x compounds affecting human health and the ecological system are NO and NO₂ (Wang et al., 2019). Photochemical processing of O₃ by NO_x has a negative impact on the atmospheric radiation budget, accounting for 10–15% of total human greenhouse radiative forcing in the atmosphere (Liu et al., 2019). NO₂ is an irritating gas that induces pulmonary oxidant harm similar to that caused by O₃ (Gordon et al., 2014). NO₂ also interacts with the hydroxyl radical (HO) that exists in the atmosphere to produce nitric acid (HNO₃), which is drained out from the atmosphere as acid rain (Kampa & Castanas, 2008). The symptoms of NO₂ are varied and include eye or nose mucosal infections, pneumonia, chronic bronchitis, pulmonary haemorrhage, and oedema (Ezratty et al., 2014).

Satellite-based measurements are now available every day for tracking atmospheric trace gases globally. Ground-based measurements are relevant for validating these space-based observations for chemical analysis, distribution, and simulation of these greenhouse gases (Wang et al., 2009). Using in situ measurements, the diurnal/seasonal variability of O₃ and NO_x for few Indian cities have been monitored (Tyagi et al., 2020; Verma et al., 2018; Yadav et al., 2014). Researchers have also explored surface O₃ and NO_x variability in India by using distinct events data — eclipse/festivals/games (Beig et al., 2013; Pakkattil et al., 2020; Saha et al., 2014). Various academics have sought to explain pollution transmission across regions in India by using back trajectory analysis and source-receptor modelling (Chandra et al., 2017; Tiwari et al., 2016). The knowledge of current changes in the time series of contaminants is utilised to reframe current regulations for an improved and healthier climate (Zeri et al., 2016). In several locations worldwide, including the Indian region, O₃ and NO_x chemistry studies have widely been carried out (Han et al., 2011; Notario et al., 2012; Pancholi et al., 2018; Tiwari et al., 2016).

In this study, we address the tropospheric levels of O₃, NO, NO₂, and NO_x from January 2015 to December 2018 in the surrounding environment of Bengaluru, Karnataka, India. The primary objectives are to investigate the diurnal and interannual monthly variations in O₃, NO, NO₂, NO_x, and their interrelationships. We have also analysed the variability of an oxidant OX (O₃+NO₂) through NO_x to understand OX's atmospheric origins in this region.

Scope of this work

India is located in a tropical region. Hence, there is intense solar radiation and high water vapour content, resulting in changes in hydroxyl radical that follows the synthesis of O₃. In the tropics, surface emissions, convection, and large-scale circulation all interact in a complex manner. Rapid urbanisation has resulted in changes in the environment. Due to increased emissions from anthropogenic sources, the concentrations of trace gases in the atmosphere vary. Simultaneous observations of O₃ and its precursors in the tropics can help researchers better understand the processes

and photochemistry of O_3 on a variety of temporal and spatial scales.

The tropical megacity of Bengaluru, India, has seen increased urbanisation over the past few decades (TERI, 2017). The increased population led to increased emissions and changed land usage patterns. All these aspects have led to changes in O_3 production and the tropospheric trace gas chemistry in this region. This study aims to understand a few aspects of that which are useful for policymakers.

Study region

Bengaluru is India's third heavily populated city, next to Mumbai and Delhi, with a populace of around 11.5 million during 2016 (TERI, 2017). The measurements were carried out at BMS College of Engineering (12.9406°N, 77.5738°E), Bengaluru. The measurements were performed to monitor atmospheric air through Air Quality Monitoring Station (AQMS) and weather parameters from an Automatic Weather Station (AWS). These were installed on the campus as part of the Ministry of Earth Sciences-funded Modelling Atmospheric Pollution and Networking (MAPAN) programme of the Indian Institute of Tropical Meteorology. Bengaluru is located at an elevation of 920 m above sea level, with uniformly distributed topography almost equidistant from both the western and eastern coasts of South India. This megacity has undergone tremendous expansion in recent years because of the massive support for information technology (IT) industry and the accompanying economic advances.

The rapid urbanisation of Bengaluru has had several urban environmental issues, including shrinking reservoirs, increased traffic jams, high levels of air pollution, flood events during heavy rains, and a rise in summer temperatures up to some extent (TERI, 2017). A study conducted by the Indian Institute of Science also found a 2–2.5 °C increase in temperature over the last decade, which has been due to 76% deterioration in vegetation and a 79% drop in water bodies (Ramachandra & Uttam, 2010). Green land surfaces that have been initially covered in vegetation are now being replaced with impervious surfaces. Such urban surfaces absorb heat energy and then re-emit it, resulting in a rise in local temperatures in urban

centres concerning surrounding suburban areas. As a result, the urban zones witness higher temperatures that lead to urban heat islands (UHI). UHI may result in changes in the local climatic zones within Bengaluru, increased consumption of energy in buildings, which may lead to elevated surface-level ozone. Over recent decades, these changes in urban morphology have increased anthropogenic heat fluxes for ozone production (Sussman et al., 2019).

Measurement techniques

Ecotech commercial analysers were used to monitor the surface level of O_3 , NO, NO_2 , and NO_x from January 2015 to December 2018. For O_3 measurements, the Serinus-10 O_3 analyser was used. The instrument operates on the principle of absorption of ultraviolet (UV) radiation at 254 nm. This analyser offers precise measurements of O_3 with a 0.5-ppbv detection limit. The machine calibration was performed periodically through an O_3 generator which is part of the analyser. Zero and span tests were also carried out to ensure the stable performance of the analyser. The nitrogen oxides were measured using a Serinus 40 NO_x analyser based on the principle of chemiluminescence. This analyser provides the mixing ratios of NO, NO_2 , and NO_x . The equipment comprises a pneumatic system, a molybdenum NO_2 -to-NO converter, a reaction cell, a detector (PMT), and electronics for processing. The detailed working processes of the O_3 and NO_x analyser are given in (Yadav et al., 2014). An automatic weather station (AWS) measures the meteorological variables such as temperature, relative humidity, wind speed, and wind direction at the measurement site.

Result and discussion

Diurnal variability of O_3

The yearly averaged O_3 diurnal variability at Bengaluru from January 2015 to December 2018 (4 years) are shown in Fig. 1a. The diurnal trend in the hourly O_3 concentration over 4-year period offers information on the production, transport, and sink mechanisms of O_3 over Bengaluru. The O_3 concentration

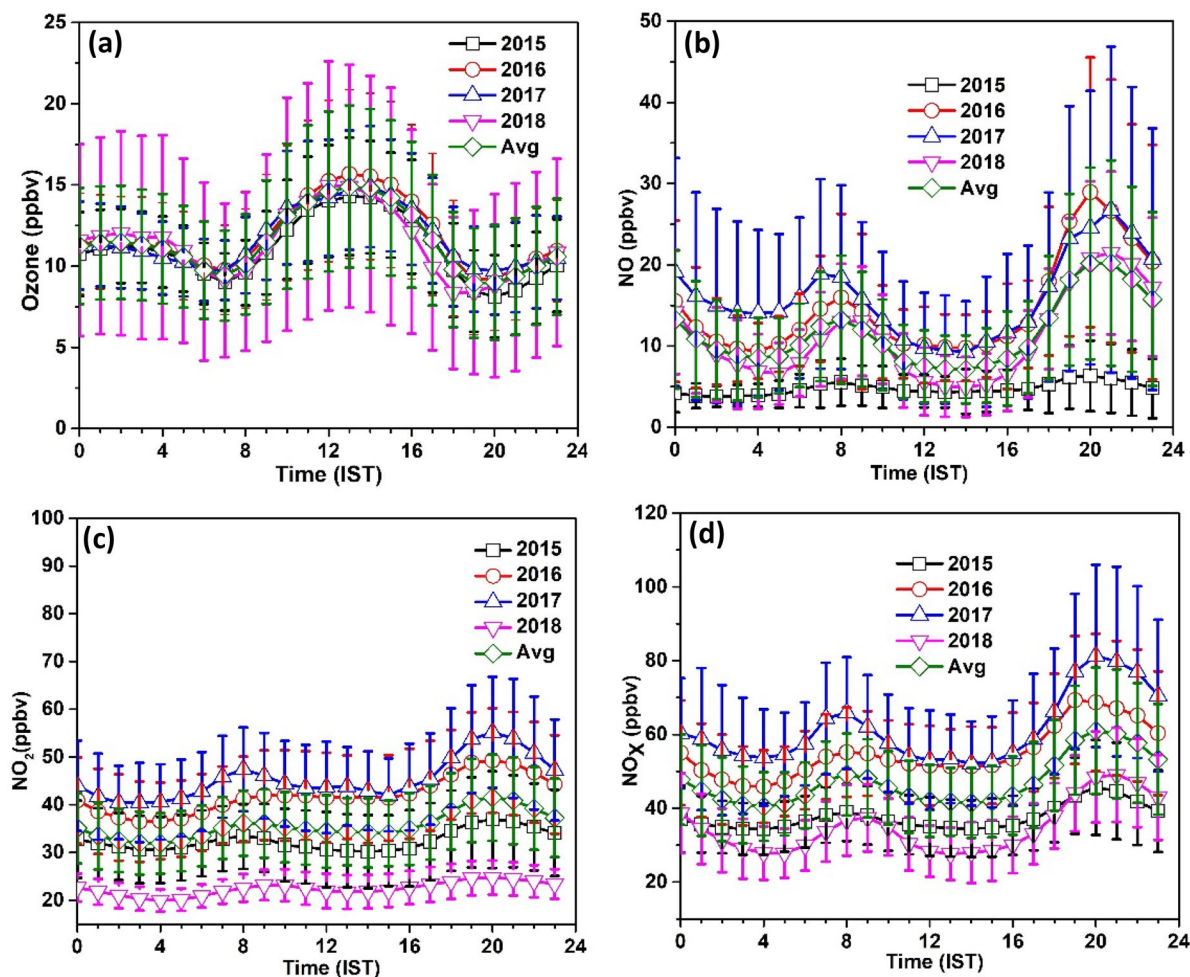


Fig. 1 a–d Diurnal variation of ozone, NO, NO₂, and NO_x

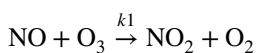
increases progressively after the photochemical output; it is highest in the daytime and then declines gradually in the evening till the next day. For all the years of observations, diel variation of O₃ is characterised by the lowest levels during early morning hours (06:00–8:00 LT) and highest levels during afternoon hours (12:00–16:00 LT), followed by minimum levels in the late evening (16:00–20:00 LT). The lower level in the early time of the day may be attributed to O₃ loss through NO and NO₂ released from photodissociation of NO₃ and N₂O₅ accumulated in the nighttime boundary layer. The highest levels are likely owing to a high mixing ratio of O₃ to precursor gases, including CO, CH₄, and TNMHC (total non-methane hydrocarbons) in the presence of NO_x and intense solar radiation in the afternoon. The

absence of NO₂ photolysis, dry deposition, and titration with NO may all contribute to the low O₃ content observed in the evening. Local climatic conditions and the diurnal pattern of the planetary boundary layer (PBL) significantly influence diurnal O₃ distributions in metropolitan areas (Saini et al., 2008). The diurnal variations of O₃ concentrations well represent the daily behaviour of urban PBLs. It usually starts with a shallow layer at night, grows into a convective structure in the morning and throughout the day, and then falls back to a shallow layer in the evening. The tropical area is defined by vigorous mixing and greater PBL due to the solar radiation and water vapour, contributing to intense convection (Yadav et al., 2014). During 2015, 2016, 2017, and 2018, the maximum O₃ concentrations over

Bengaluru were 14.3 ppbv, 15.7 ppbv, 14.9 ppbv, and 14.9 ppbv. Many studies have reported the diurnal variation of O₃ concentration at other megacities like Kanpur (2009–2013), Jodhpur (2012–2013), Kannur (2009–2011), New Delhi (2010–2012), and Chennai (May 2005 to October 2005) (Gaur et al., 2014; Nishanth et al., 2012; Pancholi et al., 2018; Tiwari et al., 2015). The observations at Bengaluru show an almost identical diurnal pattern as seen in these megacities, with minor variations in the amplitude of variability. The diurnal average daytime maximum O₃ concentrations are 14.3, 15.7, 14.9, and 14.9 ppbv, and the minimum O₃ concentrations are 8.1 ppbv, 9.1 ppbv, 9.7 ppbv, and 8.4 ppbv in 2015, 2016, 2017, and 2018, respectively at around late evening.

Diurnal variation of NO

The diurnal fluctuation of the annual mean hourly NO concentrations for several years is depicted in Fig. 1b. The diurnal fluctuation of NO exhibited two peaks, one early in the morning around 6:00–9:00 of IST (Indian Standard Time) and the other late in the evening around 19:00–21:00 LT. A valley is observed at around 11:00–18:00 LT. The highest mean concentration occurs at 8:00 LT and 20:00 LT for daytime and evening time, respectively for all the years. The maximum peak concentration during day (night) are 5.52 (6.30), 15.98 (28.94), 18.87 (26.77), and 13.05 (21.45) ppbv for 2015, 2016, 2017, and 2018, respectively. The daytime and late-night peaks match with traffic hours and hence are attributable to the same. The variation in concentrations can also be due to boundary layer processes, meteorological conditions, and emissions (Al-Jeelani, 2014). The average NO concentrations are higher in the late evening hours than in the early hours, as can be shown. Due to traffic congestion during peak hours, anthropogenic emissions are created throughout the day and a lower boundary layer height in the evening. NO concentrations fall after 9:00 LT in the morning when it combines with O₃ and oxidises to NO₂. The equation below affirms the removal of NO during the daytime



In many cities worldwide, this trend in the temporal variability of air pollutants has been observed (Han et al., 2011). Local meteorology often influences

the variations (Pudasainee et al., 2006), retaining the original trend.

Diurnal variation of NO₂

The average (\pm standard deviation) diurnal NO₂ mixing ratios measured at Bengaluru for the observation period are shown in Fig. 1c. The NO₂ mixing ratio indicates a peak between 7:00 LT and 10:00 LT in the morning hours. In the afternoon hours (12:00–16:00 LT), the minimum mixing rates were found. Later, the observation between 18:00 LT and 21:00 LT shows an increasing trend, as higher mixing ratios were seen from evening to late night. The slow increase of NO₂ concentration in the morning period of the daytime (07:00–10:00 LT) is primarily due to the increased traffic activity. Besides atmospheric stability, this is often correlated with low winds, which are the hallmark of the nocturnal boundary layer, which still exists in the early time of the day (Teixeira et al., 2009). In the afternoon, the concentration of NO₂ decreases as it correlates with the highest photolytic activity that contributes to O₃ production. The concentration of NO₂ reaches its lowest level in the afternoon, where O₃ production is at its highest. Horowitz (1982) has shown that in areas associated with strong photochemical processes, concentrations of NO₂ have exhibited a diurnal trend noticeable by a dual peak (Horowitz, 1982). Eventually, the photochemical reaction ceases after sunset, and hence, the concentration of O₃ decreases, while the concentration of NO₂ increases in the complex night-time chemistry of the atmosphere. The altitude of the mixing layer across the observation region is another factor affecting NO₂ concentrations.

The contaminants were diluted during the daytime as the mixing layer increases, while during the night-time, it was limited to within the PBL. The concentration of NO₂ begins to decrease around midnight, which may be due to the preferred conditions for converting NO₂ into N₂O₃ and N₂O₅ compared to the accumulation of NO₂ by O₃ titration. A similar diurnal variation in the city of Patna was found by Tiwari et al. (2016), which they attribute to anthropogenic emissions, local photochemistry, and dispersion condition. In Jodhpur, Pancholi et al. (2018) also showed a similar variation of NO₂ where they attribute it to vehicular emissions. Identical trends with slight differences compared to our observations in intensity

are also seen in Agra and Delhi (Sharma et al., 2010; Verma et al., 2018).

Diurnal variability of NO_x

Figure 1d shows the daily averages of NO_x for the years of the study period from January 2014 to December 2018. NO_x 's observed diurnal cycle is a double peak. The peaks are observed at 8:00–11:00 LT in the morning and 17:00–21:00 LT in the late evening. In comparison to the early morning peak, the late evening peak has a larger amplitude. The greater NO_x concentrations in the morning in Bengaluru correlate with a rise in rush-hour traffic, implying that vehicle exhaust plays a role. The decrease in NO_x concentration after 11:00 LT enhances the formation of O_3 by photolysis driven by sunlight. The pollutants are diluted during midday due to increased boundary layer height and extensive mixing. NO_x may be unable to spread vertically at night due to the reduced surface temperature, resulting in the accumulation and a high mixing ratio. In conclusion, both anthropogenic activities and boundary layer dynamics contribute to elevated NO_x levels in the early morning and late-night (Ahamed et al., 2006).

Over Bengaluru, the diurnal fluctuation of NO , NO_2 , and NO_x showed a double-peak pattern in the morning and late evening. Similar double peak patterns have been observed in other megacities, such as Ahmedabad and Kanpur, where the pattern has been related to anthropogenic activities, boundary layer dynamics, and local surface wind patterns (Gaur et al., 2014; Mallik et al., 2015). In contrast to the double peak pattern observed at Bengaluru, the observations reported by Nishanth et al. (2014) for Kannur show that the concentration of NO_x was high in the late evening hours and early morning and lower in the afternoon. Similar behaviour of elevated mixing ratios has been observed in the city of Udaipur from the evening till early morning (Yadav et al., 2014). The shallow depth of the nocturnal boundary layer resists the mixing of free tropospheric air with local pollution.

Concentrations vary according to ambient air pollution, particular emission levels, and prevailing meteorological conditions in different megacities. The appearance of double peaks in the diurnal variation of nitrogen oxides describes the study area as a significant photochemical reaction site. Nitrogen

oxides show a minimum value around the afternoon in their diurnal behaviour. The increasing intensity of solar radiation and the afternoon stretching of the mixing layer can cause the concentration of nitrogen oxides to decrease and the rate of a photochemical reaction to increase. The increase during the morning and the late-night are due to vehicular exhaust emissions and a shallow boundary layer in the night-time.

Inter annual-monthly variation of O_3

Figure 2a depicts the monthly mean of the O_3 mixing ratio from January 2015 to December 2018. It can be observed that during the summer months (March, April, May 2015), the daytime average O_3 mixing ratio was 13.19 ppbv. It follows a steady decrease in post-monsoon (October, November 2015) with the daytime mean O_3 ratio of 9.90 ppbv, 8.4 ppbv during monsoon (June–September 2015). During 2016, we see that the O_3 mixing ratio was maximum during the winter months with 14.60 ppbv, October–November with 11.87 ppbv, monsoon (June, July, August, and September) with 11.72 ppbv followed by a lower mixing ratio in March–May with 9.78 ppbv. The mixing ratio for 2017 is 15.56 ppbv during the winter months, 11.73 ppbv during the summer months, 9.1 ppbv during the monsoon months, and 12.9 ppbv post-monsoon months. The O_3 concentration for 2018 shows 9.26 ppbv (March–May) and 8.85 ppbv (June–September). Maximum O_3 concentrations occurred in March (14.43 ppbv), June (15.62 ppbv), February (16.47 ppbv), and December (36.77 ppbv), while minimum concentrations occurred in September (7.29 ppbv), April (8.23 ppbv), June (7.76 ppbv), and January (6.77 ppbv), respectively for the years 2015, 2016, 2017, and 2018. The highest concentration is linked with intense solar radiation and precursors available for the photochemical formation of O_3 during March, February, and May. The seasonal changes leading to the dry monsoon phase may account for the highest concentration observed during June 2016. The minimum concentration during September and June might be due to the monsoon period. The peroxy radicals (HO_2 and RO_2) are swept away by precipitation, restricting O_3 photochemistry in the gas phase (Seinfeld & Pandis, 2006). The lower concentration in January for the year 2018 might be due to O_3 production is slow in winter due to moderate

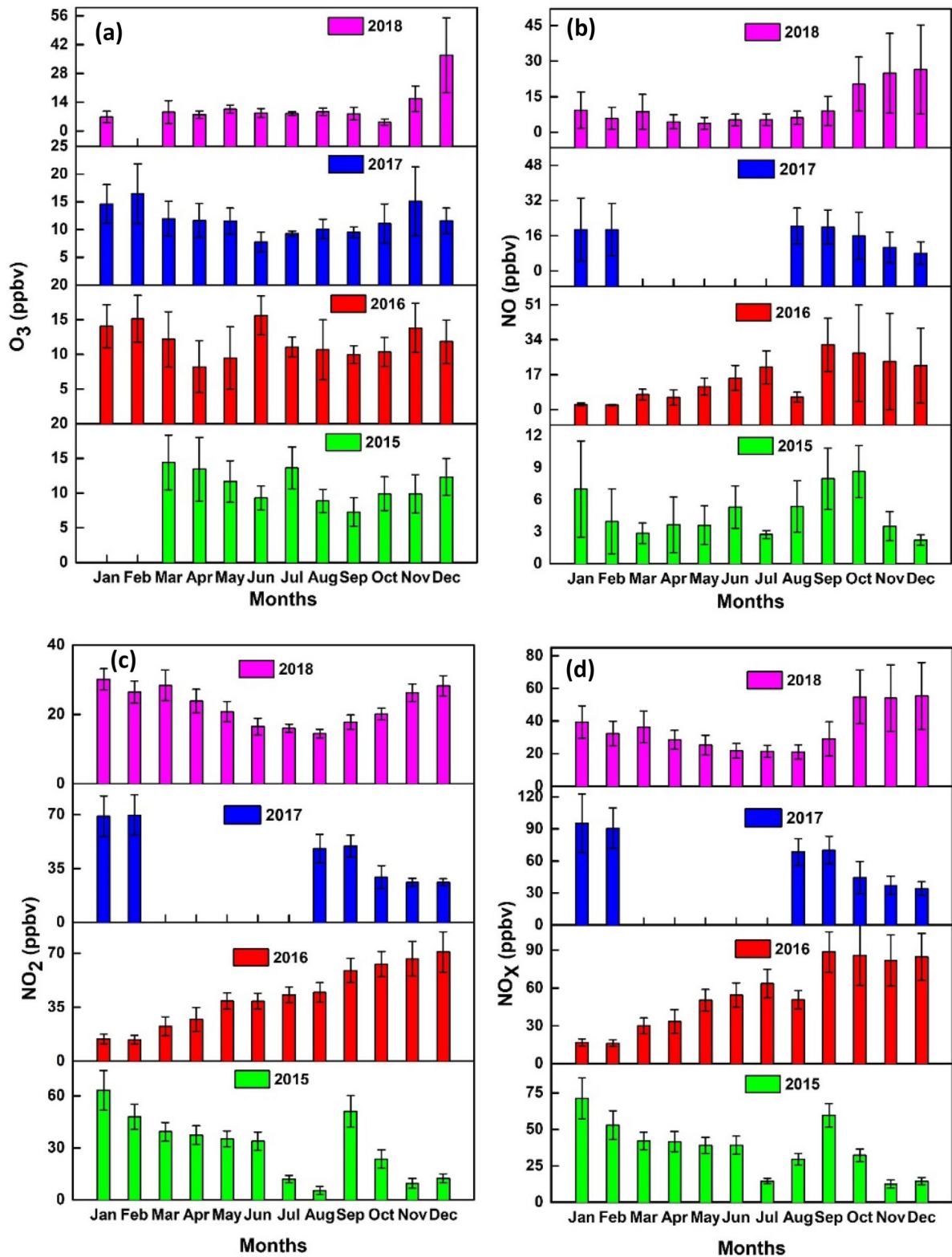


Fig. 2 a–d Monthly plots of ozone, NO, NO₂, and NO_x

solar radiation intensity. In April, a low concentration may be due to intense convection triggered by high surface temperatures.

For 2015–2018, the monthly average showed the highest concentration in February (15.8 ppbv) and the lowest concentration in September (8.75 ppbv). However, O_3 followed different monthly average variations at other sites in India. At the Udaipur, the concentration of O_3 displays extremely significant seasonality with a maximum of 28.9 ppbv in April and a minimum of 16.4 ppbv during September. Higher concentrations of CO and NO_x were associated with higher O_3 values, while lower values of O_3 were linked to lower concentrations of CO and NO_x (Yadav et al., 2014). The O_3 concentration was higher at the coastal location, Kannur, in December, while the lower (16.41 ppbv) mixing ratio was seen in July by Nishant et al. (2014). They attribute the observation to the differences in precursors and the impact of local meteorology. The overall monthly highest and lowest O_3 was seen during March (55.7 ppbv) and (16.5 ppbv) in August in Anantapur (Rama Gopal et al., 2014). Analysis revealed that the increased concentration is primarily due to intense solar radiation and the combined effect of the gaseous precursors within the mixed layer. Observations by Sharma et al. (2016) show that at Delhi, the surface O_3 mixing ratio shows high value during May, continuing with the summer (40.3 ppbv) months; the surface O_3 concentration reaches its lowest in July (16.8 ppbv). The monthly average minimum and maximum of O_3 at Agra were 14 ppbv and 57.6 ppbv, respectively. The findings in Agra by Verma et al. (2018) showed that the high value might be caused by meteorological parameters favouring the photochemical production of O_3 . The minimum levels might be due to higher cloud cover leading to lower photochemical activity and precursors scavenging. The different variations observed are due to the differences in the sources of pollution, the observation site topography, and the prevailing meteorological conditions.

The annual mean O_3 concentrations for 2015, 2016, 2017, and 2018 are 10.92 ppbv, 11.44 ppbv, 11.59 ppbv, and 8.82 ppbv. The ranges of O_3 concentrations in 2015, 2016, 2017, and 2018 are from 5.20 to 20.66 ppbv, 5.45 to 19.36 ppbv, 4.91 to 21.77 ppbv, and 1.74–18.73 ppbv, respectively. The high value may be due to meteorological conditions favouring the production of O_3 by photochemical reactions,

while the minimum value may be due to the scavenging of gases (Verma et al., 2018).

The annual average of O_3 for four years is 10.69 ppbv with a minimum and maximum varying from 5 to 24 ppbv. However, O_3 levels have fluctuated year to year in other parts of India and worldwide (Table 1). The concentrations of O_3 measured in this site are comparable to those reported in Richland, USA (Mioduszewski et al., 2011) and many cities in Malaysia (Awang et al., 2015). Compared to sites from China, Saudi Arabia and Indian sites Agra, Udaipur, Kanpur, Anantapur, Kannur, and Delhi, Bengaluru's concentration is low (Hassan et al., 2013; Nishanth et al., 2012; Rama Gopal et al., 2014; Tiwari et al., 2015; Tong et al., 2017; Verma et al., 2018; Xu et al., 2011; Yadav et al., 2016). At a semi-arid location in Anantapur (Rama Gopal et al., 2014), the annual variation of O_3 is 35.1 ppbv, with the monthly mean lowest/highest O_3 in August/March was 19.9/55.8 ppbv. The annual variance of Agra in the Indo-Gangetic plain shows the highest concentration of gaseous contaminants since pollutant emissions is higher compared to other areas of India. These annual variations can be due to the differences in the sampling site and the period of observation.

Interannual monthly variation of NO

Figure 2b shows the monthly average NO mixing ratio from January 2015 to December 2018. For 2015, the maximum and minimum monthly averages of NO were observed in October (8.25 ppbv) and December (2.22 ppbv), respectively. During 2016, maximum and minimum concentrations occurred in September (30.04 ppbv) and February (2.58 ppbv). For the year 2017, the maximum was in August (20.40 ppbv) and the minimum in December (8.06 ppbv), while for the year 2018, we see that the maximum was in December (26.47 ppbv) and minimum in April (4.45 ppbv). The occurrence of the highest concentration of NO may be attributable to anthropogenic emissions and boundary layer activities. The minimum concentrations of NO could be due to local patterns of surface wind prevailing in the city, leading to the dispersion of pollutants.

The average annual NO concentrations for 2015, 2016, 2017, and 2018 are 4.5 ppbv, 16.4 ppbv, 15.3 ppbv, and 10.7 ppbv. The lowest and the highest

concentrations of NO for 2015, 2016, 2017, and 2018 are from 1.81 to 13.09 ppbv, 1.77 to 44.63 ppbv, 4.54 to 44.57 ppbv, and 1.28 to 39.48 ppbv. The differences in NO concentration during the different periods may be attributed to the other meteorological parameters and differences in emission sources.

Table 1 presents a comparison of the annual variation of NO levels at various megacities located in different regions of India. The average mass concentration of NO is 11.72 ppbv for the 4 years of observation. As shown in Table 1, the mean NO concentrations in the current study were lower /higher or comparable to those found in other metropolises throughout the world. Bengaluru's average NO concentration was similar to that of Delhi (Tiwari et al., 2015); however, it was higher than that of Agra (Verma et al., 2018) but lower than that of Beijing (Xu et al., 2011).

Interannual monthly variation of NO₂

The monthly interannual variability of NO₂ is displayed in Fig. 2c. The monthly average NO₂ concentration during the study period for the year 2015 varies between 53.63 ppbv (January) and 5.37 ppbv (August). The maximum and minimum concentrations of NO₂ in 2016 are 70.76 ppbv (December) and 13.91 ppbv (February), and for 2017, it was observed to be 69.73 ppbv (February) and 26.06 ppbv (November). In 2018, 30.12 ppbv (January) and 14.89 ppbv (August) were the observed maximum and the minimum concentrations of NO₂, respectively. The monthly variations can be due to the collective effect of PBL dynamics and anthropogenic activities.

The yearly averaged mean NO₂ concentrations during 2015, 2016, 2017, and 2018 ranged from 2.93 ppbv to 80.8 ppbv, 0.4 ppbv to 96.8 ppbv, 5.35 ppbv

Table 1 Comparison of ozone, NO, NO₂, and NO_x with different sites

Observational place	Period of observation	O ₃ (ppbv)	NO (ppbv)	NO ₂ (ppbv)	NO _x (ppbv)	References
Ningbo, China	2012–2015	22.7			31.1	Tong et al. (2017)
Urban (Traffic)		36.1			17.8	
Suburban		37.7			12.6	
Rural						
Malaysia (Port cities)	2009	20.3		20.3		Awang et al. (2015)
Klang		15.4		12		
Perai		14.4		13.3		
Pasir Gudang						
Agia Marina, Cyprus (Rural)	1997–2012	47.5			1.8	Kleanthous et al. (2014)
Jeddah, Saudi Arabia	2011–2012	28.5–37.7			29.7–53.1	Hassan et al. (2013)
Richland, USA (Nonurban)	2007	15.4			12.7	Mioduszewski et al. (2011)
Beijing, China	1 January 2005 to 31 December 2008	24.45	26.58	29.64		Xu et al. (2011)
Urban		35.90	0.82	0.82		
Rural						
Agra, India	May 2012 to May 2013	39.6	0.8	9.1		Verma et al. (2018)
Udaipur, India	April 2011 to March 2012	22.42			11.55	Yadav et al. (2016)
Kanpur, India	June 2009 to May 2013	27.9 ± 17.8			5.7	Gaur et al. (2014)
New Delhi, India	September 2010 to August 2012	23.6	17.2	12.5	29.3	Tiwari et al. (2015)
Anantapur, India	March 2012 to February 2013	35.1	1.7	3.6	5.2	Rama Gopal et al. (2014)
Kannur, India	November 2009 to October 2010	18.4–44			2.5	Nishanth et al. (2012)
Bengaluru, India	January 2014 to December 2018	10.69	11.72	34.1	46.64	Present study

to 83.93 ppbv, 19.70 ppbv to 94.4 ppbv, and 11.58 ppbv to 35.40 ppbv, with a mean of 31.54 ppbv, 40.58 ppbv, 47.02 ppbv, and 22.38 ppbv, respectively. The annual average showed the highest concentration in January 2015, 2017, and 2018, while it showed the highest concentration in November for 2016. The minimum concentration for 2015 and 2018 was observed in August, while for 2017, it is in February, and for 2018, it is in November. The greater concentrations in January and November can be attributed to a lower mixing height, which causes pollutants to be trapped near the Earth's surface due to temperature inversion. The low NO_2 concentration may be due to the impact of changing weather parameters. The mean annual variation was highest in 2017, followed by 2016, 2015, and a minimum in 2018.

Table 1 compares the annual concentration of NO_2 at Bengaluru and the observations made at different sites. The annual mean concentration of NO_2 for the period of 2015–2018 is 34.10 ppbv. The highest and lowest concentrations during the period of study are 4 to 94 ppbv. As shown in the table, the mean NO_2 concentrations in the current study were higher/lower or comparable to those observed in other metropolises across the globe.

Interannual monthly variation of NO_x

The interannual monthly variation of NO_x is shown in Fig. 2d. In 2015 and 2016, the maximum and minimum NO_x concentrations are 66.38 ppbv (January), 86.02 ppbv (October) and 12.53 ppbv (November), and 16.22 ppbv (February), respectively. The maximum and minimum concentrations of NO_x for the year 2017 are 95.41 ppbv (January) and 34.12 ppbv (December), while the maximum concentration and minimum concentrations of NO_x for the year 2018 are 55.80 ppbv (December) and 20.99 ppbv (August). The higher concentration of NO_x during January, October, December, and lower in November, February, December, August can be attributed to meteorological parameters and the lower mixed layer depths.

For 2015, 2016, 2017, and 2018, the annual mean–variance of NO_x is 37.27 ppbv, 55.26 ppbv, 64 ppbv, and 33.99 ppbv. The minimum and maximum concentrations ranged from 7.77 to 92.31 ppbv for 2015, while it varied from 7.30 to 121.53 ppbv for 2016. It ranged from 28.6 to 120 ppbv and 11.95 to 66.077 ppbv for the years 2017 and 2018. Over the

observation period, the annual variance is due to the dynamics of the height of the PBL, which regulates the surface level of NO_x and various other pollutants, and the prevailing meteorological conditions over the measurement sites.

The annual average concentration of NO_x in Bengaluru was compared with that observed at various locations in India and worldwide (Table 1). The annual mean of NO_x at Bengaluru is 46.64 ppbv during observation years 2015–2018, with a minimum of 7.30 ppbv to a maximum of 121.53 ppbv. The NO_x concentration was comparable to that seen in a city in Ningbo, China (Tong et al., 2017), while it was higher than that observed in Kannur, Anantapur, Kanpur, Richland (USA), and Agia Marina (Cyprus) (Gaur et al., 2014; Kleanthous et al., 2014; Mioduszewski et al., 2011; Nishanth et al., 2012; Rama Gopal et al., 2014). The levels of nitrogen oxides in Bengaluru are high compared to other cities, which might be attributed to the city's rapid economic growth, industrialisation, and transportation in supporting IT and its associated industries domestically (Yang et al., 2020). The differences in the measured values of oxides of nitrogen in different locations may be attributed to the differences in location of the site, pollution sources, and local meteorological conditions.

Correlation of ozone with oxides of nitrogen

The Pearson correlation was used to analyse the link between O_3 and nitrogen oxides (NO , NO_2 , and NO_x). The precursor oxides NO , NO_2 , and NO_x all have a negative relationship with O_3 . The correlation coefficient of O_3 with NO , NO_2 , and NO_x are -0.77 ($P < 0.00$), -0.51 ($P < 0.01$), and -0.68 ($P < 0.00$), respectively. The negative correlation indicates that O_3 increases as the concentration of oxides of nitrogen decreases. It could be owing to the photochemical O_3 synthesis consuming nitrogen oxides. Furthermore, because of the high PBL, dilution of the air occurs, resulting in pollution dispersion. Photochemical generation favours O_3 formation during the daytime, while nitrogen oxides are consumed in this process. As the concentration of nitrogen oxides increases, the reaction rate of $\text{OH} + \text{NO}_2$ increases, allowing all nitrogen oxides and OH to be eliminated from the reaction cycle and reducing the formation rate of O_3 (Seinfeld & Pandis, 2006).

Pancholi et al. (2018) have reported similar results for the city of Jodhpur, where the correlation coefficient was -0.22 ($P < 0.01$) and -0.27 ($P < 0.01$) for daytime and nighttime, respectively. Mallik et al. (2015) have also reported a slope between O_3 and NO_x , which is negative. They attribute this to an incomplete photochemical reaction in the fresh air. A negative correlation with a value > -0.88 having a significance level $P = 0.01$ was observed between O_3 and NO_x in the city of Trivandrum during November 2007 to May 2009 (David & Nair, 2011). Strong negative associations have also been observed in Istanbul (Turkey), and Greater Cairo (Egypt), and their coefficients are in the range of -0.46 to -0.56 and -0.75 to -0.87 , respectively (Im et al., 2013; Khoder, 2009).

Dependence of contributions from local and regional OX

The oxidant $OX = O_3 + NO_2$ is generally used to analyse the chemical bonding between O_3 , NO , and NO_2 . Hence, this section deals with the interaction of OX and NO_x . Photochemical processes in the polluted area alter OX levels. This leads to differences in OX 's daytime and nighttime values (Mazzeo et al., 2005). The daytime (6:00–17:00 LT) and nighttime (18:00–5:00 LT) mean levels of OX and NO_x from January 2015 to December 2018 are shown in Fig. 3a–b. The total amount of OX follows the linear regression ($mx + c$) equation when plotted against

NO_x . It can be observed that the slope of the line divides OX 's total levels into a NO_x -independent and a NO_x -dependent contribution (Tiwari et al., 2015). NO_x -dependent contribution can be linked to localised pollution as a rise/fall in NO_x levels leads to a rapid increase/decrease in total oxidant levels. NO_x -independent effect can be associated with regional impact (or background factor) to the O_3 levels. Figure 3a–b show that the slope between day and night is 0.76 and 0.68, indicating that the daytime contribution is slightly higher than the nighttime contribution. Because of the effects of photochemical processes on the growth of OX , there is a difference between day and night (Mazzeo et al., 2005). The regional contributions to O_3 in daytime and nighttime, as seen by the intercept of the OX and NO_x graphs, are similar, both around 12 ppbv. In metropolitan areas, traffic emissions are the principal source of NO_x . According to research, the amount of NO_x produced as NO_2 varies depending on the type of vehicle and gasoline used (Heywood, 1988). The contribution based on NO_x depends on local emissions, and the photochemistry operating at the site depends on the local primary pollutant levels.

The variation of NO_2/OX as a function of NO_x concentration is shown in Fig. 4. The two have a clear linear relationship. There was a clear distinction between day and night. The NO_2/OX levels are almost identical during daytime and nighttime as that of NO_x . During the day and night, NO_x describes

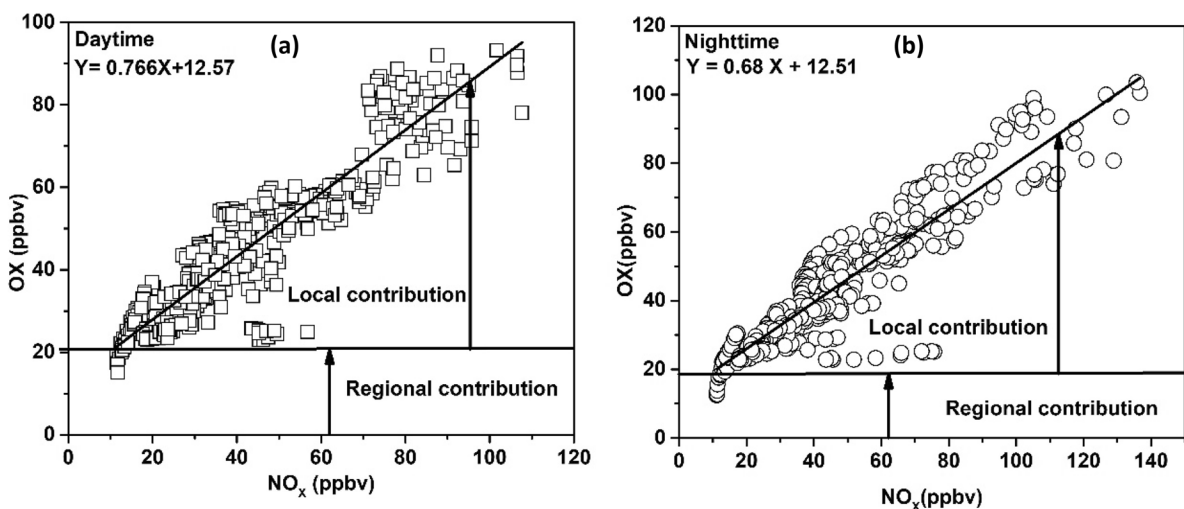
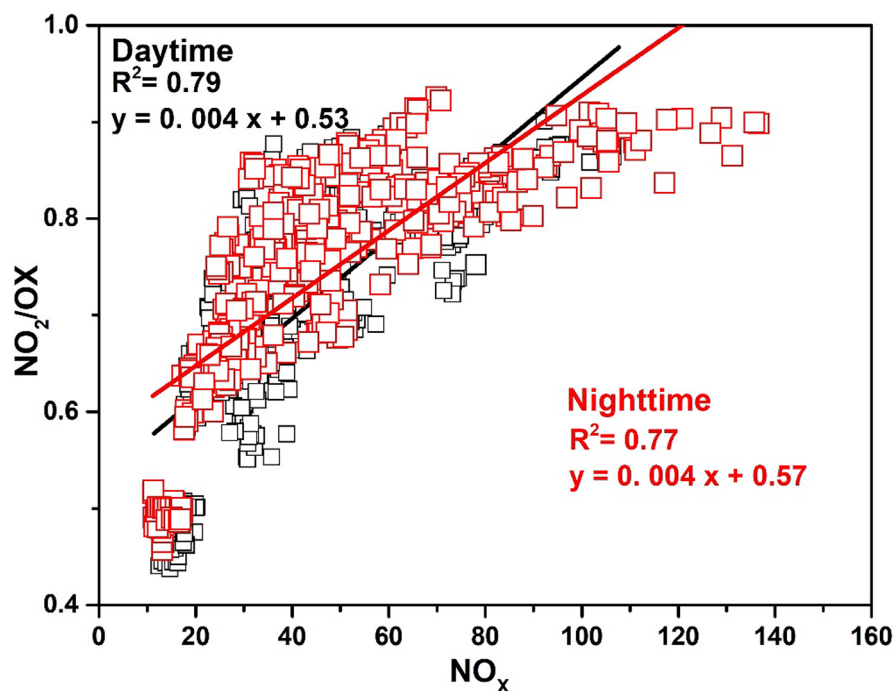


Fig. 3 a–b Variability of OX with NO_x daytime and night-time

Fig. 4 Variation of OX with NO_2/OX ratio



79% and 77% of the NO_2/OX difference. The NO_2/OX and NO_x slope is the same during the day and night, which is 0.004. During day and night, the local NO_x levels remain almost constant. Due to photolysis reactions, the local contribution of NO_x throughout the day equals out with the build-up of NO in late-night transport events; the total local NO_x concentrations remain consistent during the day. In Tianjin, China, and Riyadh, Saudi Arabia, related variations are noted (Han et al., 2011; Shareef et al., 2018). NO_x has been identified as a crucial source in the development of O_3 in other places, including Kathmandu, Nepal, and Delhi, India (Pudasainee et al., 2006; Tiwari et al., 2015).

Conclusion

In a metropolitan region of Bengaluru, Karnataka, India, continuous in situ observations of O_3 , NO, NO_2 , and NO_x were made between January 2015 and December 2018. O_3 has a unimodal peak in its diurnal distribution. The peak showed the highest built-up of O_3 in the noontime (12:00–16:00 LT) that was influenced due to increased photochemical creation of O_3 , which its precursors produce in

the presence of solar radiation. The diurnal distribution of nitrogen oxides showed a bimodal peak, with one during the morning and the other during the late night that can be associated with traffic density. The monthly interannual variability of O_3 and nitrogen oxides showed distinct variation due to differences in anthropogenic emissions, PBL dynamics, and meteorological parameters. The variance in O_3 and NO_x could be attributable to changes in land use and land cover resulting from urbanisation over Bengaluru. An assessment of the variability of OX ($\text{O}_3 + \text{NO}_2$) with NO_x is done to comprehend the oxidative processes of the urban environment. In this respect, the OX vs NO_x was plotted; the slopes indicate the local contribution, while the intercept indicates the regional contribution. The results of the slope showed that local effects were more significant in the daytime than the nighttime. The observed variations may be due to photochemical reactions involving the formation of OX. Such long-term continuous monitoring of ozone and its precursors and meteorological parameters are critical for better understanding the distribution of air pollutants at various sites, especially metropolitan regions in developing countries like India.

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Data availability Data that support the results of this study are available from the corresponding author upon reasonable request.

Declarations

Disclaimer The thoughts and conclusions expressed in this research study may not reflect those organisations with which the authors collaborate.

Conflict of interest The authors declare no competing interests.

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