Ozone chemistry and dynamics at a tropical coastal site impacted by the COVID-19 lockdown

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The nationwide lockdown in India to curb the spread of Coronavirus disease 2019 (COVID-19) led to colossal reduction in anthropogenic emissions. Here, we investigated the impact of lockdown on surface ozone (O3) and nitrogen dioxide (NO2) over a tropical coastal station – Thumba, Thiruvananthapuram (8.5°N, 76.9°E). Daytime as well as night-time NO2 showed reduction by 0.8 (40%) and 2.3 (35%) ppbv, respectively during the lockdown period of 25–30 March 2020 as compared with the same period of previous 3 years. Unlike many urban locations, daytime surface O3 is found to be dramatically reduced by 15 ppbv (36%) with O3 production rate being lower by a factor of 3 during the lockdown. Interestingly, a feature of O3-hump during the onset of land breeze typically observed during 1997–1998 has reappeared with magnitude of 5–10 ppbv. A photochemical box model, capturing this feature, revealed that significant O3 sustained till onset of land breeze over the land due to weaker titration with NOx during lockdown. It is suggested that the transport of this O3 rich air with onset of land breeze led to the observed hump. Our measurements unravel a remarkable impact of the COVID-19 lockdown on the chemistry and dynamics of O3 over this tropical coastal environment.

Keywords. Thumba; trace gases; photochemistry; land and sea breeze.

1. Introduction

After the outbreak of Coronavirus disease 2019 (COVID-19) pandemic, lockdowns were imposed in various countries to minimize the spread. The lockdowns have been shown to drastically reduce the anthropogenic emissions improving the air quality significantly (Bauwens et al. 2020; Le et al. 2020). Large reductions in nitrogen dioxide (NO2) over major cities have been observed worldwide (Dantas et al. 2020; Liu et al. 2020). Globally, the reduction in tropospheric O3 burden by about 6 Tg (~2%) is suggested to have altered the radiative forcing significantly (Miyazaki et al. 2020). The major source of NO2 is fossil fuel burning in vehicles, industries, power generators, etc. NO2 is one of the major air pollutants and also the main precursor of ozone (O3; Seinfeld and Pandis 1998). O3 plays a key role in tropospheric chemistry as the major source of hydroxyl radical (OH), which is known as detergent for removal of many pollutants from the atmosphere. O3 also controls the oxidation capacity of the troposphere, besides its role as a greenhouse gas. O3 is not directly emitted in the
atmosphere, rather it is formed through photochemical reactions, involving the precursor gases such as nitrogen oxides \((NO_x = NO + NO_2)\), CO, and hydrocarbons (Seinfeld and Pandis 1998). Two competing chemical processes, \(O_3\) production via photolysis of \(NO_2\) vs. \(O_3\) destruction by NO, govern \(O_3\) variability in urban and rural environments. While higher \(O_3\) levels during daytime are due to dominance of photochemical production, the titration reaction dominates during night-time causing lower \(O_3\) levels (Seinfeld and Pandis 1998). Nevertheless, the dependence of \(O_3\) on the concentrations of the precursors is complex and non-linear. Therefore, accurate measurements over distinct environment are necessary to characterise the chemical regimes and to understand role of various atmospheric processes.

A nationwide lockdown was also imposed by the Government of India starting since 25 March 2020 (https://mha.gov.in/sites/default/files/PR_Nation aLockdown_26032020_0.pdf). A 14-hr voluntary ‘Janata (i.e., public) curfew’ was also observed on 22 March 2020. The lockdown was implemented in a phased manner, during phase-1 (25 March–14 April 2020) and phase-2 (15 April–03 May 2020), the imposed guidelines restricted movements of all vehicles, industrial activities, and other services, except the essential and emergency services. Thus, anthropogenic emissions of trace gases and aerosols from various sources such as vehicles, industries, etc., largely came to a halt. Since phase-3 (04 May 2020 onwards), certain relaxations were implemented which included vehicular movements and a few other anthropogenic activities. However, lockdown restrictions remained stringent over the hotspot regions (https://www.mha.gov.in/notifications/circulars-covid-19). Recently, a few studies have reported the reduction of air pollutants near source regions over megacities in India (Kumari and Toshniwal 2020; Mahato et al. 2020; Sharma et al. 2020) and synoptic changes over the Indian region (Shehzad et al. 2020). Black carbon concentrations showed reductions in the range of 16–60% with respect to climatological mean over the Indian region (Gogoi et al. 2021). Observations from 134 monitoring sites of Central Pollution Control Board across India showed reduction of \(\sim 40–70\%\) in \(PM_{2.5}\), \(\sim 40–60\%\) in \(PM_{10}\); \(\sim 30–70\%\) in \(NO_2\); and \(\sim 20–40\%\) in CO, but the changes in surface \(O_3\) were observed to be site-specific (Dhaka et al. 2020; Singh et al. 2020; Soni et al. 2021). However, \(O_3\) chemistry over tropical coastal environments away from strong sources but impacted by land–sea breeze systems, has not yet been explored. In this regard, we have investigated the changes in surface \(O_3\) as well as \(NO_2\) over a tropical coastal station by analysing \textit{in-situ} measurements during the first week of lockdown (25–30 March 2020). In addition, measurements available over India’s capital, Delhi (28.7°N, 77.1°E) were used to infer the heterogeneity in surface \(O_3\) chemistry.

2. Experimental site, instrumentation and data

The observational site Thumba, Thiruvananthapuram (8.5°N, 76.9°E, \(\sim 3\ m\ amsl\)), is situated on the southwest coast of India, approximately 500 m away from the Arabian Sea coast. This tropical coastal site experiences typical mesoscale circulation comprising of sea-breeze (SB) during daytime and land-breeze (LB) during night-time almost throughout the year (Narayanan 1967). Thiruvananthapuram district has a population of 3.3 million as per census 2011.

\textit{In-situ} measurements of surface \(O_3\) and \(NO_x\) were performed using ultraviolet (UV) photometric \(O_3\) analyser (model 49 C) and chemiluminescence \(NO_x\) analyser (Model 42i) of Thermo Electron Corporation, USA, respectively. Based on absorption of UV radiation at \(\sim 253.7\ nm\) by \(O_3\) molecules, \(O_3\) mixing ratios in sample air were derived using the Beer–Lambert law. \(NO_x\) analyser works on the principle of chemiluminescence property of \(NO_2\) molecules. The oxidation of \(NO\) by \(O_3\) molecules forms excited \(NO_2\) molecules. The measurement of luminous radiation emitted by these excited \(NO_2\) molecules is a measure of \(NO\) concentration. \(NO_2\) is measured by converting it into \(NO\) using molybdenum converter under high temperature. Sample air was drawn from \(\sim 3\ m\) above the ground using a Teflon tube and pre-conditioned air (dehumidified and filtered for dust particles) was fed to the analysers for these measurements. \(O_3\) analyser was calibrated with an in-built \(O_3\) generator and a zero air generator. The uncertainty in \(O_3\) mixing ratio, lower detection limit, and linearity are \(\pm 5\%\), 1 ppbv and 1\%, respectively. \(NO_x\) analyser was calibrated using standards traceable to the National Institute of Standards and Technology diluted with custom-made calibrator. Lower detection limit, linearity, and uncertainty for \(NO_x\) measurements are 0.05 ppbv, 1\% and \(\pm 5\%\), respectively (Tanimoto et al. 2007). Data were recorded at 5 min intervals. Further details of instruments as well as site description can be found
elsewhere (Girach et al. 2012; Nair et al. 2018). The observations during 25–30 March 2020 (6 days), besides observations during pre-lockdown period and previous years are used in the present study.

In addition, surface wind speed and direction used in the study were measured using the automatic weather station (AWS), Dynalab Weathertech Pvt. Ltd, India, having accuracy of ±0.5 ms⁻¹ and ±3°, respectively.

For the purpose of comparison of O₃ changes, measurements at 14 different sites across Delhi (28.7°N; 77.1°E) made under the National Air Quality Monitoring Programme (NAMP) by Central Pollution Control Board (CPCB) under Ministry of Environment, Forest and Climate Change (MoEFCC) of India are also used. Hourly averaged O₃ data falling beyond 3-sigma standard deviations were filtered prior to estimation of daily averages. Details on measurements and filtering criteria can be found in the CPCB reports (CPCB 2011) and previous studies (Schnell et al. 2018).

3. Results and discussions

Diurnal patterns of surface O₃ is characterised by higher mixing ratios during daytime and lower mixing ratios during night-time, opposite to NO₂ diurnal variations, over Thumba. These variations in O₃ and NO₂ are significantly influenced by photochemistry, SB and LB activity, besides the impact of boundary layer changes (Nair et al. 2002; David and Nair 2011; Girach et al. 2012). Daytime (12–17 hrs) and night-time (23–06 hrs) average NO₂ variations are shown in figure 1 during 19–30 March 2020 and during the same period of previous 3 years (during 2016–2019). Generally, night-time NO₂ level is higher than that of daytime (figure 1) due to absence of NO₂ photolysis, titration of O₃ with NO producing NO₂, shallow boundary layer height limiting the vertical mixing, and prevalence of LB transporting NOₓ rich air to the study site (David and Nair 2011; Girach et al. 2012). Daytime NO₂ during Janata curfew day showed some reduction, and daytime NO₂ value of the previous day is comparable to that of the mean value of this day of previous years. However, reduction in night-time NO₂ was beyond 1-sigma variability due to reduced emission during daytime under the curfew restrictions. A quick response to reduction in emission is mainly due to a shorter lifetime of NO₂ (2–8 hrs; Liu et al. 2016). Although unexpected, NO₂ remained lower on subsequent day. Daytime and night-time NO₂ during lockdown period of 25–30 March 2020 were observed to be significantly lower (beyond 1-sigma standard deviation) with respect to mean values in previous years. Average values during this period are shown by horizontal dashed lines in figure 1. While NO₂ was reduced by 0.8 ppbv (40%) during daytime, reduction during night-time is seen to be 2.3 ppbv (35%). Larger reduction in absolute magnitude of night-time NO₂ is expected as LB transports the relatively polluted airmass from the land. Since night-time NO₂ levels are associated with daytime O₃ (David and Nair 2011), impact on daytime O₃ is expected to be significant as discussed subsequently. The observations from Tropospheric Monitoring Instrument (TROPOMI) aboard Sentinel-5 Precursor satellite showed 1.0×10¹⁵ ± 3.0×10¹⁴ molecules cm⁻² tropospheric NO₂ during 24–30 March 2020 over the study location, which was lower by ~40% as compared to the level (1.7×10¹⁵ ± 1.3×10¹⁴ molecules cm⁻²) during same period of previous year. This reduction was also beyond the 1-sigma standard deviation and in line with the percentage reduction seen in daytime surface NO₂.

Figure 2(a) shows the daytime O₃ during lockdown period (yellow shaded region) and the same period of previous years. Daytime O₃ showed
reduction by 15 ppbv (36%), however, small reduction of \( \sim 4 \) ppbv in night-time \( O_3 \) was not significant (within standard deviation; see supplementary figure S1). These observations of daytime \( O_3 \) reduction are in contrast with studies from other urban environments showing daytime enhancement (Resmi et al. 2020; Soni et al. 2020; Panda et al. 2021). Figure 2(b) shows the mean diurnal variations of \( O_3 \) where suppression in \( O_3 \) production is clearly evident during lockdown period. Considering the \( O_3 \) during morning hours (07–10 hrs), estimated slope from linear regression analysis is a measure of \( O_3 \) production rate. \( O_3 \) production rate which was 9.8 ppbv h\(^{-1}\) during previous years is found to be reduced to 3.3 ppbv h\(^{-1}\), i.e., by a factor of 3 during the lockdown. Figure 2(c) shows the mean diurnal variations of \( NO_2 \) where effective suppression of fumigation peak during morning hours is clearly evident, besides lower values during day and night. Since \( O_3 \) production rate depends on precursor concentrations, lower levels of \( NO_2 \) (and other precursors) in the morning hours (figure 2c) could result in reduced production rate and lower daytime \( O_3 \). It is worth to mention here that \( NO_2 \) dispersed over the sea during LB (night-time) gets recirculated, after chemical losses, during SB which prevails during daytime over the site (David and Nair 2011). Lower \( NO_x \) during night could have enhanced \( O_3 \) by reducing its titration with NO during night-time (23–06 hrs). However, since \( O_3 \) level itself was reduced during daytime, influence of reduced titration is compensated and significant change in night-time \( O_3 \) was not observed. However, the slower evening loss in \( O_3 \) was observed as discussed subsequently.

SB component (SBC) is calculated as \([\text{wind speed} \times \sin(325° – \text{wind direction})]\) as the coastline of Thiruvananthapuram is aligned along \( \sim 145–325° \). Figure 3 shows diurnal variations of \( O_3 \) for three individual days (25–27 March 2020) along with SB component. SBC is used to demarcate SB and LB, as positive and negative values denote SB and LB, respectively. Crossing from SB to LB denote the onset of LB as marked by red dashed line in figure 3. In normal conditions, it has already been illustrated that \( O_3 \) remains high till the onset of LB (David and Nair 2011; Girach et al. 2012). \( O_3 \) started decreasing with reduction in solar radiation (sunset time is marked with black dashed line in figure 3). Interestingly, a hump of 5–10 ppbv around the LB onset is observed over three days. This feature was observed consistently during 1997–1998 over the site as reported by Nair et al. (2002). However, it has not been observed regularly in the recent years, as reported by David and Nair (2011).

In order to explain the \( O_3\)-hump with onset of LB, we have performed simulations using a photochemical box model. We used Master Mechanism
model – version 2.5, developed at National Center for Atmospheric Research (NCAR), which simulates chemical evolution of air parcel with time considering the detailed photochemistry in absence of dynamics or further emissions. The detailed description of the model can be found elsewhere (Madronich 2006; Nair et al. 2011; Ojha et al. 2012; Soni et al. 2021). The environmental conditions such as latitude, longitude, date, parcel elevation, total column ozone, SO\textsubscript{2} column, NO\textsubscript{2} column, aerosol optical depth, single scattering albedo, aerosol angstrom coefficient, temperature, air density were set to typical values of 8.5\textdegree N, 76.9\textdegree E, 26 March 2020, 10 m, 280 DU, 0.12 DU, 0.11 DU, 0.50, 0.82, 1.2, 300 K, 2.45\texttimes 10\textsuperscript{19} molecules cm\textsuperscript{-3}, respectively as obtained from Ozone Monitoring Instrument (https://disc.gsfc.nasa.gov/), ground-based observations at the study site and literature (Babu et al. 2007; David and Nair 2011). The initial/background concentrations of carbon monoxide, methane, isoprene, formaldehyde, ethane, ethene, propane, propene, \textit{n}-butane, and \textit{i}-butane were included as 300, 2000, 1, 0.7, 2.5, 5, 1.5, 3, 5, and 0.1 ppbv, respectively, which represent the typical coastal/rural conditions. The background value of O\textsubscript{3} has been set as free tropospheric value of 40 ppbv (Ajayakumar et al. 2019). The crucial diurnal constraints used in the simulation were the observed nitric oxide (NO) and NO\textsubscript{2} (supplementary figure S2). In order to account for the overestimation in the measurement of NO\textsubscript{2} due to molybdenum convertor (Winer et al. 1974), observed NO\textsubscript{2} were used in the simulation after giving an offset of 35\% based on sensitivity simulations. In addition, variation of boundary layer height and entrainment velocity were assumed to their typical values of 0.2–1.5 km and 0.05–20 cm s\textsuperscript{-1}, respectively (Bremaud et al. 1998; Girach et al. 2012).

We performed first simulation for airmasses sampled at Thumba site based on above-mentioned model configuration. As SB airmass travels over the land during daytime, it gets enriched by precursors (i.e., NO\textsubscript{2}) due to, for instance, vehicular emission. Thus, by giving a small increment of 35\% in NO\textsubscript{2}, we performed another simulation for airmasses over land. As Thumba site experiences airmasses from land during night-time, we have combined the two simulations in figure 4. The model reproduced the observed diurnal feature of O\textsubscript{3}, including the O\textsubscript{3}-hump around onset of LB. However, magnitude of simulated O\textsubscript{3}-hump is ~5 ppbv, smaller than the observed value, which could be partly due to discount of transport. Diurnal variation of O\textsubscript{3} simulated for the airmass over land showed higher levels of O\textsubscript{3} during daytime, which is in agreement with the observations at an inland station Kariavattom (8.56\textdegree N; 76.89\textdegree E, ~4 km from the coast), which is close to Thumba (supplementary figures S3 and S4). Significant O\textsubscript{3} level sustained in the evening hours due to lower NO caused by reduced emission under lockdown. This slow losses of O\textsubscript{3} after sunset, results in significant O\textsubscript{3} even in evening hours, similar to that over a rural site in India (Naja and Lal 2002). Thus, in nutshell, this feature of O\textsubscript{3}-hump is attributed to

Figure 3. Diurnal variations of O\textsubscript{3} (black) and sea breeze component for 3 days during lockdown period. Black and red dashed lines represent sunset time and onset of land breeze, respectively.

Figure 4. Observed and model simulated diurnal variation of surface O\textsubscript{3} on 26 March 2020. Yellow shaded region demark the SB airmass. Blue dots connect O\textsubscript{3} simulated for airmasses over land and sea.
the weaker $O_3$ titration with NO over land during lockdown. This allowed a significant amount of $O_3$ to sustain over land and get transported to the observational site with the onset of LB.

It is worth mentioning that under normal condition (unlike lockdown), higher NO in the evening hours titrates $O_3$ effectively after sunset and $O_3$ levels in the LB are much lower and hence $O_3$-hump is not seen under normal condition. The above mechanism would also explain the $O_3$-hump reported earlier (Nair et al. 2002) due to lower NOx at that time (Nair et al. 2018).

To compare the $O_3$ behaviour in a contrasting environment, we utilised the $O_3$ observations over NOx-rich environment, Delhi. Figure 5 shows the daytime $O_3$ averaged over 14 monitoring stations across Delhi during lockdown period of 25 March–04 April 2020. After the implementation of lockdown surface $O_3$ showed reduction by $\sim 25$ ppbv; however, after a few days, $O_3$ started increasing slowly converging to reference curve of 2019 afterwards. The increase in $O_3$ across Delhi (Mahato et al. 2020) and over another urban location, Ahmedabad (Soni et al. 2021) was mainly attributed to stronger diminish of NO which titrates $O_3$, non-linear chemistry and changes in meteorology. Thus, overall daytime $O_3$ reduction is insignificant over Delhi specifically few days after lockdown. This feature shows the non-linear and complex $O_3$ chemistry for urban locations where plenty of volatile organic compounds are present besides heterogeneous chemistry under significant aerosol loading. This also points to the contrasting behaviour of chemistry and dynamics of $O_3$ as observed over a coastal station.

4. Summary and conclusions

Present study highlights the impact of lockdown on surface $O_3$ chemistry over a tropical coastal station, Thiruvananthapuram, during first week of lockdown (25–30 March 2020). $O_3$ precursor, NO2 reduced significantly (2.3 and 0.8 ppbv during night-time and daytime, respectively) which led to suppression of observed $O_3$ production rate by a factor of 3. Suppression of fumigation peak in NO2 contributed in lowering the $O_3$ production rate. Consequently, daytime (12–17 hrs) surface $O_3$ reduction of 15 ppbv (36%) was seen. Interestingly, surface $O_3$ showed a hump (5–10 ppbv) around the onset of land breeze. The $O_3$-hump was reproduced by the simulations using a photochemical box model. Reappearance of this feature which was seen during 1997–1998, is due to a weaker titration slowing the evening time $O_3$ losses over the land and sustaining significant $O_3$ levels till the onset of land breeze. This study shows that the lockdown has influenced the $O_3$ chemistry and dynamics dramatically over this tropical coastal region.

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Author statement

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Figure 5. Mean daytime $O_3$ over Delhi during lockdown period of 2020 and during the same period of 2019. Error bars represent standard deviations. Yellow shaded region and red arrow denote the lockdown period and $O_3$ reduction during the initial days of lockdown, respectively.
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