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Integrated process analysis retrieval of changes in ground-level ozone and fine particulate matter during the COVID-19 outbreak in the coastal city of Kannur, India

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The Community Multi-Scale Air Quality (CMAQ) model was applied to evaluate the air quality in the coastal city of Kannur, India, during the 2020 COVID-19 lockdown. From the Pre1 (March 1–24, 2020) period to the Lock (March 25–April 19, 2020) and Tri (April 20–May 9, 2020) periods, the Kerala state government gradually imposed a strict lockdown policy. Both the simulations and observations showed a decline in the PM2.5 concentrations and an enhancement in the O3 concentrations during the Lock and Tri periods compared with that in the Pre1 period. Integrated process rate (IPR) analysis was employed to isolate the contributions of the individual atmospheric processes. The results revealed that the vertical transport from the upper layers dominated the surface O3 formation, comprising 89.4%, 83.1%, and 88.9% of the O3 sources during the Pre1, Lock, and Tri periods, respectively. Photochemistry contributed negatively to the O3 concentrations at the surface layer. Compared with the Pre1 period, the O3 enhancement during the Lock period was primarily attributable to the lower negative contribution of photochemistry and the lower O3 removal rate by horizontal transport. During the Tri period, a slower consumption of O3 by gas-phase chemistry and a stronger vertical import from the upper layers to the surface accounted for the increase in O3. Emission and aerosol processes constituted the major positive contributions to the net surface PM2.5, accounting for a total of 48.7%, 38.4%, and 42.5% of PM2.5 sources during the Pre1, Lock, and Tri periods, respectively. The decreases in the PM2.5 concentrations during the Lock and Tri periods were primarily explained by the weaker PM2.5 production from emission and aerosol processes. The increased vertical transport rate of PM2.5 from the surface layer to the upper layers was also a reason for the decrease in the PM2.5 during the Lock periods.

1. Introduction

Tropospheric ozone (O3) and fine particulate matter (PM2.5) are of significant concern due to the adverse effects on air quality, agriculture, climate, and human health. India, the world’s second most populated country, has experienced rapid deterioration in air quality due to industrialization and economic growth over the past decades (Karambelas et al., 2018; Nishanth et al., 2012). High tropospheric ozone concentrations in India were found to be less conducive to plant production in the most important cultivation area (Oksanen et al., 2013). A study revealed the lower loss for rice crops as compared with wheat mainly attributed to lower surface ozone levels during the crop season after the Indian summer monsoon (Lal et al., 2017). The potential health risk caused by exposure to these pollutants has also aroused extensive attention in India. In 2011, about 570,000 premature mortalities occurred due to exposure to PM2.5 and 12,000 people died of chronic obstructive pulmonary disease (COPD) owing to O3 exposure on a national scale in India (David et al., 2019). Estimated annual

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premature mortality attributed to long-term exposure to ambient PM$_{2.5}$ exceeded 1 million in 2015 in India (Cohen et al., 2017).

Severe Acute Respiratory Syndrome Corona Virus-2 (SARS-CoV2), officially classified as COVID-19, spread throughout the world and was declared a global pandemic by the World Health Organization (WHO) on March 11, 2020 (Al-Qahtani, 2020). In India, the first confirmed COVID-19 case was identified in the state of Kerala on January 30, 2020 (Gautam and Hens, 2020). Later, COVID-19 gradually became prevalent in Maharashtra, Gujarat, Delhi, and the rest of the states in India (Naqvi et al., 2021). To mitigate the spread of COVID-19, the Prime Minister of India imposed a nationwide lockdown as a precautionary measure for 21 days beginning on the 24th of March (Kumari and Toshniwal, 2020). Some of the lockdown measures included the enforcement of home quarantine, the shutting down of academic institutes, offices, industries, and markets, and the limitation of public transport to ensure social distancing (Goel, 2020; Ravindra et al., 2021). Recent studies have reported air quality improvements in India during the lockdown period were experienced due to the reduced transportation and economic activities (Gautam, 2020; Karuppasamy et al., 2020; Pant et al., 2020; Ramasamy and Adlaya, 2020). Goel (2020) discovered a significant improvement in air quality during the COVID-19 lockdown with reductions of 60%, 40%, and 30–40% in PM$_{2.5}$, NO$_{x}$, and O$_{3}$ concentrations, respectively, as compared to the same period during the previous two years. Gouda et al. (2021) reported reductions in NO, NO$_{x}$, NO$_{2}$, SO$_{2}$, PM$_{2.5}$, and O$_{3}$ concentrations of 47.3%, 49%, 49%, 10%, 37.7%, and 15.6%, respectively, during the lockdown (as compared to the pre-lockdown period). Air quality improvement was also observed in West Bengal, with a reduction of 58.71%, 57.92%, and 55.23% in the PM$_{2.5}$, PM$_{10}$, and NO$_{2}$ concentrations, respectively, from the pre-lockdown phase to the lockdown phase (Sarkar et al., 2020). Even in the megacity of Delhi, both the nationwide lockdown and the city-scale restriction were responsible for improving air quality; particulate matter concentrations and the average air quality index (NAQI) both saw dramatic reductions when compared to previous years or the pre-lockdown period (Kumar et al., 2020; Mahato and Pal, 2022; Mahato et al., 2020; Pal et al., 2022; Pandey et al., 2021; Singh and Kumar, 2021). Inferring from several studies that have focused on the whole of India or different regions of India, distinct disparities were found in the changes in air quality between the different areas (Biswal et al., 2021; Dave et al., 2021; Kaluri et al., 2021; Pathakoti et al., 2020; Saxena and Raj, 2021). It was concluded that the improvement in air quality caused by the lockdown measures seemed to be not obvious as people expected in some locations, and even appeared to be negative (Chen et al., 2020; Ghosh et al., 2020; Manchanda et al., 2021; Meng et al., 2021; Sbai et al., 2021b; Tibrewal and Venkataraman, 2022; Wu et al., 2021; Yin et al., 2021; Zoran et al., 2020). Mor et al. (2021) found that the SO$_{2}$, O$_{3}$, and m,p-xylene concentrations continued to increase throughout the study period in Chandigarh. Agarwal and Kumar (2022) observed that the CO concentration was greater in 2020 than in the corresponding time in 2019 over Wazirpur, Delhi. Biswal et al. (2021) demonstrated NO$_{2}$ observations enhancements of up to approximately 25% during the lockdown associated with fire emissions over northeastern and central India. In particular, the phenomenon of elevated ozone levels during the lockdown periods has been extensively reported (He et al., 2021; Meng et al., 2021; Sbai et al., 2021b; Wu et al., 2021; Yin et al., 2021). Previous studies have analyzed the reasons for O$_{3}$ changes from the perspective of emission restrictions and meteorological factors including relative humidity, temperature, solar radiation, and ozone-forming mechanisms (Meng et al., 2021; Mor et al., 2021; Ren et al., 2021; Saxena and Raj, 2021; Tibrewal and Venkataraman, 2022; Tobias et al., 2020). An increase in air temperature and a rise in solar radiation was responsible for the O$_{3}$ increase during the lockdown because higher temperatures could decrease the stability of the atmosphere and correspondingly increase the mixing height of pollutants, and stronger solar radiation could have enhanced the intensity of photochemical reactions in the atmosphere (Dang and Liao, 2019; Mor et al., 2021; Ravindra et al., 2019; Saxena and Raj, 2021; Sbai et al., 2021b). The PM$_{2.5}$ reduction during the lockdown would cause more infiltration of solar radiation through the atmosphere and retard the photochemical reaction that forms ozone (Saxena and Raj, 2021; Sbai et al., 2021b; Zoran et al., 2020). O$_{3}$ reacts with nitrogen oxide (NO) and is degraded by the titration process as follows: NO + O$_{3}$ = NO$_{2}$ + O$_{2}$ (Akimoto and Tanimoto, 2022; Gronoff et al., 2019; Kumari and Toshniwal, 2020). Several studies conducted in different areas of India have attributed the O$_{3}$ increment during the lockdown to the reduced O$_{3}$ consumption caused by decreased NO levels (Kalluri et al., 2021; Selvam et al., 2020; Tibrewal and Venkataraman, 2022). Allu et al. (2021) investigated the impact of the lockdown on the air quality in Hyderabad City and revealed an increase in O$_{3}$ concentrations from 26 ppb (pre-lockdown) to 56.4 ppb (lockdown) that was related to a decrease in CO and NO$_{x}$ concentrations. Estimates of the air quality in the western region of India by Nigam et al. (2021) also verified a rapid decline in most of the pollutant concentrations (PM$_{10}$, PM$_{2.5}$, CO, and SO$_{2}$) and an increase in the ozone concentration due to a significant decrease in NO$_{2}$ (by 80.18%) during the lockdown. Kumari and Toshniwal (2020) found that the O$_{3}$ concentration increased by 37.35% compared with the pre-lockdown phase in Delhi, and this was due to a decrease in the NO levels that reduced O$_{3}$ consumption and consequently resulted in an O$_{3}$ increase. O$_{3}$ enhancements attributed to the lockdown-derived NO$_{x}$ emission reduction and the lower O$_{3}$ titration by NO were observed in some recent studies, not only in India, but also in different parts of China (Meng et al., 2021; Wu et al., 2021; Yin et al., 2021), France (Sbai et al., 2021b), Italy (Sicard et al., 2020), and Spain (Tobias et al., 2020).

These studies typically interpreted the O$_{3}$ increase based on an analysis of the ozone observations or described the observed trends of other pollutants during the lockdown. However, the evolution of air pollutants is a complex process that involves a variety of physical processes, such as emissions, deposition, advection, and diffusion coupled with a chemical process and so on (Huang et al., 2005; Jeon et al., 2012; Wang et al., 2014; Xu et al., 2008). Even though previous studies have reported the air quality changes during COVID-19 lockdown and identified the main driving forces of the changes, few studies could quantify the changes from different driving forces and individual processes. Therefore, in this study, we quantitatively elucidated the contributions from different processes to changes in PM$_{2.5}$ and O$_{3}$ during the lockdown to improve our understanding of the complex interactions between chemical and physical processes of air pollution. It is hard to identify the contributions of chemical and physical processes behind the air quality change induced by the lockdown measures solely based on the observation data since limited measurements are available. Numerical models deal with the temporal and spatial changes of air pollutants and simulate the formation processes by solving a set of partial differential equations that may help us to clarify the limited understanding of physical and chemical processes. The reasons for air quality changes observed during the lockdown period in India have not yet been investigated adequately, and only limited model studies have been reported. Some examples of air quality models used to investigate the effect of lockdown on air quality include the WRF-CHIMERE model (Drumka et al., 2020) and the WRF-CMAQ model (Zhang et al., 2021). Hence, it is necessary to employ numerical simulations to further investigate the above-mentioned processes and understand the causes of the air quality changes during the COVID-19 lockdown.

In this study, the Community Multi-Scale Air Quality (CMAQ) (Byun and Schere, 2006) model was applied to investigate the temporal and spatial variations of PM$_{2.5}$ and O$_{3}$ during the lockdown in the west coast city of Kannur in Kerala State, India. The CMAQ has been widely utilized to evaluate air quality and can provide robust support for numerical simulations of various air pollutants (Hu et al., 2017; Hu et al., 2015; Liu et al., 2020; Wang et al., 2021a). In addition to the national lockdown period that began March 24, the Kerala state government imposed a “triple-lockdown” (more tightened restrictive measures compared to the lockdown period) in Kannur beginning April 20 for 20 days, as Kannur
was identified as a “hotspot” of COVID-19 and a “red” zone in the state. More details regarding the lockdown and “triple-lockdown” measures have been explained elsewhere (Resmi et al., 2020). To clarify the reasons for the increasing trends in the O₃ concentrations and the descending trends in the other air pollutants during the lockdown and “triple-lockdown” period, the integrated process rate (IPR) analysis embedded in the CMAQ was employed to evaluate the contributions of individual atmospheric processes, such as gas-phase chemistry, dry deposition, cloud processes, aerosol processes, emissions, vertical transport, and horizontal transport.

2. Methods

2.1. Study area

The study area was Kannur city located in the north of Kerala State in southwestern India (Fig. 1). Kannur, with a land area of about 3000 km², is the sixth-largest urbanized area in Kerala and is known for its high levels of literacy and healthcare (Nishanth et al., 2011; Resmi et al., 2020). The land of Kerala can be divided into three natural areas of low land, middle land, and high land, each of which is almost parallel from north to south (Sheela et al., 2017). The high land consists of mountains stretching in eastern Kerala, forming a natural wall with an average altitude of 1 km, separating Kerala from the adjoining States. The middle land, characterized by undulating terrain, is rich in agricultural products like paddy, tapioca, banana, pepper, ginger, and areca nut. The low land area is flat and consists of strips running along the coast of the Arabian Sea. The landmass in Kannur city mainly consists of middle land and low land (Sheela et al., 2017).

Kerala is classified as a tropical wet climate under the Koppen classification system and as warm humid under the Indian climatic zone map (Joshima et al., 2021). The climate in Kerala is divided into four seasons namely winter (December–February), summer (March–May), monsoon (June–August), and post-monsoon (September–November) (Nishanth et al., 2014). The summer season is marked by high convective movement and intense sunlight. At Kannur, the temperature is high from March to May and is low from June through August. The wind direction changes slowly towards the southwest during summer with slightly higher wind speeds of around 11–12 km/h (Ct et al., 2021).

2.2. Data collection

Observation data on O₃, PM₂.₅, and other tracer pollutants (PM₁₀, NO, NO₂, CO, and SO₂) were collected from 1st March to May 9, 2020 and reported in Resmi et al. (2020). The observational site is in Kannur town (11.87° N 75.37° E 3 m MSL) in the northern part of Kerala state, which lies in a coastal belt along the Arabian Sea and is very close to the National Highway (NH 66). The study period was divided into three periods to study the impact of lockdown on the variation of trace pollutants over Kannur. The collected hourly observations have been further calculated in an individual period. Thus, daily average concentrations and periodical average diurnal variations of tracer pollutants in three spans were available.

2.3. Model setup and input data

The CMAQ model version 5.2 was applied with the SAPRC07 gas-phase photochemical mechanism (Carter, 2010) and AERO6 aerosol reaction mechanism (Binkowski and Roselle, 2003) to reproduce the air quality in the coastal city of Kannur, India, during the COVID-19 lockdown in 2020. The CMAQ model, developed as the 3rd generation of the comprehensive air quality model by the U.S. Environmental Protection Agency, provides a state-of-the-art representation of the processes that affect the fate of airborne pollutants. The CMAQ model has been widely used in air pollution numerical studies (Hu et al., 2016; Hu et al., 2015; Li et al., 2021; Sharma et al., 2020; Shi et al., 2020; Zhang and Ying, 2012; Zhang et al., 2016; Zhang et al., 2021). As shown in Fig. 1, the model was configured with a simulation domain covering Southwest India and parts of the Arabian Sea with a horizontal resolution of 4 km × 4 km (87 × 87 grid cells). The blue dot denotes the location of Kannur town (11.87° N 75.37° E). The modeling period was divided into three periods similar to Resmi et al. (2020); namely a pre-lockdown period (abbreviated as the Pre1 period) of 24 days (March 1–24, 2020), a lockdown period (abbreviated as the Lock period) of 26 days (March 25–April 19, 2020), and a triple-lockdown period (abbreviated as the Tri period) of 20 days (April 20–May 9, 2020). The vertical structure of simulation was based on a sigma coordinate system, among which a total of 18 vertical layers were distributed corresponding to sigma levels of 1.000, 0.996, 0.990, 0.980, 0.970, 0.960, 0.950, 0.940, 0.930, 0.910, 0.880, 0.850, 0.775, 0.690, 0.600, 0.475, 0.290, 0.090, and 0.000 at the boundaries of the layers. A spin-up of 2 days was used to minimize the influence of initial conditions (IC).

Meteorological fields were generated using the Weather Research & Forecasting model (WRF) version 4.2 with National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Global Analysis data. The data came from the U.S. National Center for Atmospheric Research (NCAR), with a spatial resolution of 1.0° × 1.0° (https://rda.ucar.edu/datasets/ds083.2/). The Regional Emission inventory in Asia (REAS) version 3.1 (Kurokawa and Ohara, 2020) collected the emissions of sulfur dioxide (SO₂), NOx, carbon monoxide (CO), PM₁₀, PM₂.₅, black carbon (BC), organic carbon (OC), NH₃, carbon dioxide (CO₂), and non-methane volatile organic Compounds (NMVOCs) species, which could be projected to 12 sources (domestic, extraction, fertilizer, industry, manure management, misc, other transport, power plants point, power plants non-point, road transport, solvents, waste) to characterize the anthropogenic emission in India. The biogenic emissions were estimated by using the Model for Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012).

2.4. Model evaluation protocol

Meteorological parameters obtained from National Climate Data Center (NCDC), including relative humidity (RH), the temperature at 2 m above the ground level (T2), wind speed (WS), and wind direction (WD) at 10 m were compared with the simulated data (ftp://ftp.ncdc.noaa.gov/pub/data/noaa/). As Table S2 depicts, the performance was
validated by calculating several statistical metrics, including mean bias (MB), mean error (ME), and root mean square error (RMSE) using the following equations, which were suggested by Emery et al. (2001):

\[
MB = \frac{1}{N} \sum_{i=1}^{N} (P_i - O_i) 
\]

(1)

\[
ME = \frac{1}{N} \sum_{i=1}^{N} |P_i - O_i| 
\]

(2)

\[
RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (P_i - O_i)^2} 
\]

(3)

where \(N\) represents the total number of the data; \(P_i\) is the ith predicted value; \(O_i\) is the ith observed value.

The simulations of air quality during different periods were evaluated against observations performed by Resmi et al. (2020) over Kannur during the COVID-19 outbreak. The statistical indexes of normalized mean deviation (NMB), normalized mean error (NME), and correlation coefficient (\(r\)) were defined by equations (4)–(6) to evaluate the predicted air quality, which was suggested by Emery et al. (2001).

\[
NMB = \frac{\sum_{i=1}^{N} (P_i - O_i)}{\sum_{i=1}^{N} O_i} \times 100\% 
\]

(4)

\[
NME = \frac{\sum_{i=1}^{N} |P_i - O_i|}{\sum_{i=1}^{N} O_i} \times 100\% 
\]

(5)

\[
r = \frac{\sum (P_i - \bar{P}) \times (O_i - \bar{O})}{\sqrt{\sum (P_i - \bar{P})^2 \times (O_i - \bar{O})^2}}
\]

(6)

where \(N\), \(P_i\), and \(O_i\) are consistent with that mentioned earlier. \(\bar{P}\) and \(\bar{O}\) indicate the average value of all observed and predicted data, respectively.

### 2.5. The integrated process rate analysis

Process analysis (PA) is a versatile analytical technique for separating and quantifying the contributions of individual physical and chemical processes to the changes in the predicted concentrations of a pollutant. The CMAQv5.2 model was equipped with a PA module to solve the physical and chemical processes involved in \(O_3\) and \(PM_{2.5}\) formation and other selected model output species. As one of the two components in PA analysis (including Integrated Process Rate (IPR) analysis and Integrated Reaction Rate (IRR) analysis), IPR analysis deals with hourly change in species concentration which attributes to gas-phase chemistry, dry deposition, cloud processes, aerosol processes, emissions, vertical advection and diffusion, horizontal advection and diffusion for each grid cell in the model domain. The cloud process represents the net effect of cloud extinction, cloud scavenging, aqueous phase chemistry, mixing of chemical species under and in cloud, and wet deposition. The aerosol process represents the net effect of aerosol thermodynamics, new particle generation, condensation of gas (H\(_2\)SO\(_4\), HNO\(_3\)) and organic carbon on preexisting particles, and coagulation between particles of different modes (Zhang et al., 2019). The IPR method has been widely applied in identifying the relative importance of individual atmospheric processes (Li et al., 2012; Liu et al., 2010; Wang et al., 2010; Xu et al., 2008). The principle of IPR is a problem of solving mass continuity equations. A more detailed description of the solution to the mass equations was discussed by (Huang et al., 2005).

In this paper, the results of IPR were employed to perform the process analysis involved in \(O_3\) and \(PM_{2.5}\) formation at the surface layer and in the planetary boundary layer (PBL) at the grid cell of Kannur, respectively. For the IPR analysis of \(O_3\) chemistry (gas-phase), dry deposition, cloud processes, horizontal transportation (including advection and diffusion), and vertical transportation (including advection and diffusion) were considered. Various atmospheric processes such as emissions, aerosol processes, chemistry, dry deposition, cloud processes, horizontal transportation, and vertical transportation acted together for the formation and evolution of \(PM_{2.5}\) pollution. According to the influence of \(O_3\) or \(PM_{2.5}\) concentration, atmospheric processes can be divided into two categories; source process (increase in concentration, a situation where IPR >0) and sink process (reduction in concentration, a situation where IPR <0). Emission and dry deposition belong to the source process and sink process respectively; The IPR of the chemistry process, aerosol process, cloud process, horizontal transportation, and vertical transportation can either be positive or negative. The contribution of individual atmospheric processes in the generation of \(O_3\) and \(PM_{2.5}\) can be calculated by the following formula.

\[
SOURCE_p = \sum_{t}^{p} IPR_{p,t} \times 100\% \text{ (IPR}_{p,t} > 0) 
\]

(7)

\[
SINK_p = \sum_{t}^{p} IPR_{p,t} \times 100\% \text{ (IPR}_{p,t} < 0) 
\]

(8)

where \(p\) represents the atmospheric process, and \(t\) represents the hour. \(SOURCE_p\) represents the proportion of the atmospheric process \(p\) in all source processes, \(SINK_p\) represents the proportion of the atmospheric process \(p\) in all sink processes. Both can reflect the importance of this atmospheric process in affecting the changes in species concentrations.

### 3. Results and discussion

#### 3.1. Emission adjustment

Previous studies have demonstrated anthropogenic emission reductions during the COVID-19 lockdown (Fioletov et al., 2021; Huang et al., 2021; Zheng et al., 2020; Zheng et al., 2021). Liu et al. (2021) found the decline of \(NO_2\) tropospheric vertical column density (VCD) values between the pre- and peri-period in 2020 was considerably sharper than the same periods in past years, which means a significant reduction in \(NO_2\) emission during the lockdown in India. A drastic lockdown induced a decline in nitrogen dioxide emissions was also reported in the Indian cities of Delhi and Mumbai (Shehzad et al., 2020). Sikarwar et al. (2021) claimed a 14.3% drop in the total global \(CO_2\) emissions with a maximum contribution from the transportation sector (58.3%) for January through April 2020 compared to the year before. Beig et al. (2021) reported a reduction of 66.39% and 62.14% in \(CO\) and \(SO_2\) emission during the lockdown over Delhi and Mumbai in India, respectively. The hypothesis about the reduction of anthropogenic emissions used in model simulation during the COVID-19 pandemic has also been mentioned elsewhere (Jiang et al., 2021; Zhang et al., 2021). Considering a massive reduction in vehicular movement, industrial and other activities caused by tightened restrictive measures during the Lock and Tri periods as mentioned above, we assumed that anthropogenic emissions should also be reduced according to the strictness of the movement restrictions to better reproduce the trends in air pollution during the COVID-19 pandemic.

#### 3.2. Validation of model performance

Table 1 depicts the statistical summaries of the trace pollutants (\(O_3\), \(PM_{2.5}\), \(PM_{10}\), \(NO\), \(NO_2\), \(CO\), and \(SO_2\) over Kannur compared with the observation-based concentrations reported in Resmi et al. (2020). For \(O_3\), the simulated results were comparable to the observations, as the
NMB and NME values during the Pre1, Lock, and Tri periods all met the benchmarks. The NMB value during Pre1 was negative, suggesting an underestimation of the modeled O
defined mean bias; NME is normalized mean error; r is correlation coefficient). The model performance benchmarks were suggested by (Emery et al., 2017).

Table 1
Model performance of O
during Pre1, Lock, and Tri periods all met the benchmarks, and the NMB values during all the simulated data during the different periods (indicated by All in Table 1), the NMB value was slightly overestimated but also within the benchmark. In general, the PM
during the Pre1, Lock, and Tri periods exhibited a similar pattern, with the peak in O
during the daytime, the higher temperatures and more abundant sunlight provided a better environment for photochemical reactions in the atmosphere, which resulted in enhanced O
dept observed concentrations during the predicted periods, except during the Pre1 period. Although the NMB value during Pre1 exceeded the benchmark, the NMB and NME values during the other periods all satisfied the benchmarks, and the NMB values of all the model predictions for PM
during the Pre1 period to the Tri period. The average predicted concentrations of the daily PM
during the lockdown were consistent with the evolution of field observations (Resmi et al., 2020). Similar changes were also reported by Naqvi et al. (2021) and Saxena and Raj (2021).

The diurnal variations of O
during the Pre, Lock and Tri periods exhibited a similar pattern, with the peak in O
during between 1200 and 1400 local standard time (LST), while the concentration was lower in the night and early morning hours. During the daytime, the higher temperatures and more abundant sunlight provided a better environment for photochemical reactions in the atmosphere, which resulted in enhanced O
during the COVID-19 lockdown.

Fig. 2 displays a comparison between the simulated and observed concentrations of the daily averages in O
during the Pre1, Lock, and Tri periods. It was observed that the simulated O
during the COVID-19 outbreak, as reported by Zhang et al. (2021). The average predicted concentrations of daily O
during the Pre1, Lock, and Tri periods were 23.50, 29.57, and 28.36 ppb, respectively (the observed values were 23.69, 25.62, and 26.78 ppb, respectively). This indicates an enhancement in the O3 concentration despite various activities being cut off due to the COVID-19 lockdown. Although there was a gap between the simulated and observed PM, both values were found to have declined considerably from the Pre1 period to the Tri period. The average predicted concentrations of the daily PM
during the lockdown were 47.84, 34.94, and 20.95 μg/m³, respectively, compared with the observed concentrations of 72.04, 48.73, and 28.1 μg/m³, respectively. In summary, the increase in O3 and decrease in PM
during the lockdown were consistent with the evolution of field observations (Resmi et al., 2020).

In Fig. 2, the observed daily average O3 and PM2.5 are shown in Fig. 3. The model simulations reproduced the diurnal variations and magnitude of O3 well over Kannur. The diurnal variations of O3 during the Pre, Lock and Tri periods exhibited a similar pattern, with the peak in O3 appearing between 1200 and 1400 local standard time (LST), while the concentration was lower in the night and early morning hours. During the daytime, the higher temperatures and more abundant sunlight provided a better environment for photochemical reactions in the atmosphere, which resulted in enhanced O3 formation (Chen et al., 2019; Mor et al., 2021; Wang et al., 2022; Yang et al., 2020; Yin et al., 2019).

Fig. 3. Periodical average diurnal variations of O3 and PM2.5 during the Pre1, Lock, and Tri periods at Kannur. The Pre period consists of the Pre1 and Pre 2 (May 10–17, 2020) periods.
addition, an increase in the air temperature can decrease the stability of the atmosphere and correspondingly increase the mixing height, leading to an increase in the vertical mixing of \( \text{O}_3 \) in the troposphere (Akpınar et al., 2008; Ravindra et al., 2019). It should be noted that the daily average concentrations and peak of \( \text{O}_3 \) during the Lock and Tri periods were higher than that during the Pre1 period, indicating an increase in the \( \text{O}_3 \) concentrations even during a strict lockdown period. As exhibited in Fig. 3, the model captured the daily trend of PM\(_{2.5}\), but with a certain degree of underestimation throughout the entire period. Compared with the other two periods, PM\(_{2.5}\) during the Pre1 period exhibited a stronger diurnal variation with a relatively higher concentration in the morning and lower concentration during the afternoon hours. The diurnal variations in the other two periods were not well pronounced, but the PM\(_{2.5}\) concentration also showed a downward trend from morning to afternoon. The PM\(_{2.5}\) concentrations were found to decrease significantly from the Pre1 period to the Tri period for both the simulated and observed results. Comparisons of the predicted and observed concentrations further demonstrated that the \( \text{O}_3 \) and PM\(_{2.5}\) formation were captured reasonably well over Kannur.

Fig. 4 demonstrates the spatial distribution of predicted air pollutant concentrations during the Pre1 period and changes in concentration between the Lock period and the Pre1 period, as well as the Tri period and the Pre1 period. High \( \text{O}_3 \) concentrations in southwest India mainly appeared in the central and northern regions of the domain, spread across the three south Indian states (Kerala, Karnataka, and Tamil Nadu). Compared with Pre1 days, the \( \text{O}_3 \) enhancement during the Lock period mainly appeared along the coast of the Arabian Sea, covering most areas of North Kerala. While during the Tri period, except in the coastal areas, the \( \text{O}_3 \) increase was also found in South Karnataka and North Tamil Nadu. \( \text{O}_3 \) levels in other areas mostly dropped during the COVID-19 outbreak. Prior to the lockdown, PM\(_{2.5}\) concentrations were mainly concentrated in Kerala. It was clear from Fig. 4 (PM\(_{2.5}\) panel) that the PM\(_{2.5}\) concentrations during the COVID-19 outbreak mainly decreased in the study domain. The variability of regional distribution was also reported by Zhang et al. (2021) and Naqvi et al. (2021). Fig. S1 shows the spatial distributions of other air pollutants (PM\(_{10}\), NO, NO\(_2\), CO, and SO\(_2\)). From Fig. S1, the reductions in air pollution concentration were observed during the Lock and Tri periods. In general, \( \text{O}_3 \) concentration showed an upward trend, while PM\(_{2.5}\) showed a downward trend over Kannur during the COVID-19 outbreak.

### 3.3. IPR analysis for ozone formation

Fig. 5 reveals the hourly contributions of the individual atmospheric processes to the formation of \( \text{O}_3 \) and the hourly concentrations of \( \text{O}_3 \) during the three periods. As shown in Fig. 5a, the vertical transport from the upper layers dominated the surface \( \text{O}_3 \) formation during each period, contributing 89.4%, 83.1%, and 88.9% of the total \( \text{O}_3 \) during the Pre1, Lock, and Tri periods, respectively (the contribution ratios are shown in Fig. 7). Although photochemistry could have affected the \( \text{O}_3 \) concentrations negatively or positively, it primarily played a critical role in \( \text{O}_3 \) formation.

Fig. 5. Contributions of the individual processes to the concentrations of \( \text{O}_3 \) (a) at the surface layer and (b) in the planetary boundary layer during the three periods, where CHEM, DDEP, HTRA and VTRA, and CONC denote \( \text{O}_3 \) change by gas-phase chemistry, reduction in \( \text{O}_3 \) by dry deposition, change in \( \text{O}_3 \) by horizontal and vertical transportation, and the hourly \( \text{O}_3 \) (in ppb) respectively.

Fig. 4. Spatial distributions of predicted \( \text{O}_3 \) and PM\(_{2.5}\) concentrations during the Pre1 period and changes between the Lock period and the Pre1 period, as well as the Tri period and the Pre1 period.
removal during the three periods, with the highest negative contribution ratio of 47.1% during the Pre1 period and the lowest ratio of 35.21% during the Tri period. Horizontal transport and dry deposition processes were the other two major sinks of O₃, accounting for 52.2–62.7% of the O₃ sinks during the three periods. The O₃ concentrations at the surface layer during the Lock and Tri periods exceeded the O₃ concentration during the Pre1 period. Compared with the Pre1 period, O₃ enhancement during the Lock period was primarily attributed to the lower negative contribution of the photochemistry processes rather than O₃ titration by higher NOx emissions as during the Pre1 period. The lower O₃ removal rate by horizontal transport also accounted for a considerable portion of the O₃ increase during the Lock period. For the Tri period, gas-phase chemistry also exhibited a slower consumption of O₃ as compared to the Pre1 period, during which more O₂ was removed due to titration by higher NOx emissions. Another possible reason for the higher O₃ levels during the Tri period was that the upper boundary layer had a stronger vertical transport import of surface O₃ than that during the Pre1 period.

From the perspective of the entire planetary boundary layer (Fig. 5b), the interpretation of concentration changes during the COVID-19 outbreak was slightly different. The O₃ enhancement during the Lock and Tri periods were both related to the increased O₃ vertical transport. For the Tri period, a weaker photochemical removal still made sense for the O₃ increase, while the contributions of photochemistry during the Pre1 and Lock periods were comparable. It was concluded that contributions of the individual atmospheric processes varied in the upper layers above the ground. The mean hourly O₃ change rates due to various atmospheric processes for layers 1 to 10, as well as the evolution of the O₃ vertical profiles during the three periods, are shown in Fig. S2. At surface layer 1, the titration rate of O₃ by NOx was 10.55 ppb/h, 8.96 ppb/h, and 7.86 ppb/h during the Pre1, Lock, and Tri periods, respectively. The contributions of gas-phase photochemistry were all positive in the other upper layers except the first two layers, achieving the highest positive value at layers 4–5 during the three periods. The dry deposition of O₃ only occurred at the surface layer. For layers 1–10, horizontal transport predominately contributed to O₃ removal, and the removal rate generally decreased with increasing height. The evolution of the vertical transport contribution was more varied in the vertical layers, showing a general trend of negative values at the upper layers and positive values at the lower layers. The mean hourly O₃ concentrations during all three periods increased with the added vertical layers, indicating high O₃ formation in the upper layers. The strong vertical gradient of O₃ concentrations between the lower and upper layers causes vertical O₃ transport from the upper to the lower (Huang et al., 2005). Hence, O₃ formation in the upper layers was predominantly attributed to strong gas-phase photochemistry, and O₃ import at the surface layer primarily originated from vertical transport.

Fig. 6 illustrates the diurnal variation for contributions of the different processes to O₃ formation during the three periods. As shown in Fig. 6a, for the Lock and Tri periods, the surface O₃ concentrations remained at a low level prior to 0700 LST, and the O₃ levels gradually accumulated. The positive effect of vertical transport became obvious, subsequently approaching peaks of 62.2 ppb and 53.7 ppb at 1300 and 1400 LST, respectively. During the buildup of the daytime maximum O₃, the O₃ removal rate via dry deposition and horizontal transport increased considerably, whereas vertical transport served as the primary contributor to compensate for the loss and enhanced O₃ levels. The obvious contributions of vertical transport during this period were associated with the increasing air temperature, which directly decreases the stability of the atmosphere and correspondingly increases the vertical mixing height of O₃ (Alpinar et al., 2008; Mor et al., 2021; Ravindra et al., 2019). Later, the O₃ concentration declined under the synergistic effect of the weakened vertical import, the continuous negative effect of dry deposition, horizontal transport, and gas-phase chemistry, until 1800 LST. Gas-phase photochemistry exhibited the consumption of O₃ during most times of the day, except between 1000 and 1500 LST. In the early hours of the morning, photochemistry is a vital process to remove surface O₃ due to titration reactions caused by intensive local NOx emissions. During the daytime, especially during 1000–1500 LST, when ozone precursors and solar radiation are both sufficient, the photochemical effect tends to be positive, thus generating surface O₃ (Li et al., 2012). Horizontal transport displayed a negative effect on the net O₃ during the daytime and a positive effect in the nighttime, which might have been due to the local air circulation over the Kannur coast that was characterized by sea breezes in the daytime and land breezes at night. In general, O₃ input from the upper layer was the primary source of near-surface O₃ throughout the day during the Lock and Tri periods, with the highest positive contributions of 50.8 ppb/h and 57.4 ppb/h (compared with 48.11 ppb/h during the Pre1 period). The diurnal evolution of the O₃ concentrations during the Pre1 period shared similar trends with the Lock and Tri periods, though there was a slight delay. The O₃ levels during the Pre1 period increased rapidly from 0800 LST, showing an hour delay in contrast with 0700 LST during the Lock and Tri periods. Additionally, the O₃ peak appeared 1–2 h later than that during the lockdown. It is worthy to note that the delay in O₃ concentrations was generally in good agreement with the delay in the vertical transport contributions, and this further confirmed the dominant influence of vertical transport on the surface O₃ input over Kannur. During the Pre1 period, the photochemical titration by high NOx emissions was stronger, and the photochemical O₃ generation was weaker than those during the other two periods, making the biggest average negative effect of photochemistry appear during the Pre1 period relative to the Lock and Tri periods.

Photochemistry made a small contribution to the surface O₃ formation; nevertheless, it showed a greater contribution to O₃ generation in the entire boundary layer (as shown in Fig. 6b), especially during the daytime. Because of the increase in solar radiation, sunshine and temperature can promote ozone formation during the daytime (Chen et al., 2019; Liu et al., 2019; Mor et al., 2021; Wang et al., 2022; Yang et al., 2020; Yin et al., 2019). In addition, rising temperature enhances the biogenic volatile organic component (BVOC) and volatile organic component (VOC) emissions. Their oxidation produces ozone that contributes to an increase in O₃ (Lee and Kim, 2012; Pugh et al., 2013; Sbai et al., 2021b; Song et al., 2019; Wang et al., 2021c). In terms of the entire planetary boundary layer, the positive effects of photochemistry during the Lock and Tri periods lasted longer in an entire day and exhibited higher peaks than those during the Pre1 period, which was more conducive to the accumulation of ozone in the boundary layer. During
the nighttime, the contributions of vertical transport in the boundary layer became the major O\textsubscript{3} import. The more pronounced positive effects of nighttime vertical transport during the Lock and Tri periods could have accounted for the O\textsubscript{3} increase relative to the Pre1 period. Horizontal transport served as an ozone removal pathway during most times of the day except for several hours at night. The effect of horizontal transport was relatively negligible in the entire boundary layer.

In summary, a significant increase in O\textsubscript{3} formation during the COVID-19 lockdown was caused by a lesser NO\textsubscript{x} titration effect (due to NO\textsubscript{x} emission reductions) during the nighttime and more active photochemical generation during the daytime, and this result adequately exhibited the weaker negative effect of photochemistry during the day compared with the Pre1 period. The increase in O\textsubscript{3} and reasons for O\textsubscript{3} changes during the lockdown were compared between this study and previous studies in Table 2. The average O\textsubscript{3} increase (23.3\%) for Lock and Tri days compared to Pre1 days was slightly lower than 29\% stated by Kumari and Toshniwal (2020), lower than 37\% reported by Dumka et al. (2021), and below the range of 25–48\% reported by Selvam et al. (2020), while exceeded the change reported by Kalluri et al. (2021) and Mahato et al. (2020). These discrepancies highly depended on the different study areas and the duration of the lockdown periods. O\textsubscript{3} enhancement during the COVID-19 lockdown resulting from less NO\textsubscript{x} titration has been proposed previously over other regions such as cities in China (Meng et al., 2021; Ren et al., 2021; Wu et al., 2021).

Table 2

| Study Site                              | Study Period          | Main Findings                                                                 | Reasons for air pollution changes                                                                 | References                  |
|-----------------------------------------|-----------------------|-------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------|-----------------------------|
| Kannur City                             | Lock: 3.25–4.19; Tr: 4.20–5.9 | O\textsubscript{3} increase (+25.8\%); PM\textsubscript{2.5} (−27\%);                   Less O\textsubscript{3} titration; weaker emission and aerosol processes; transport process | Present Study                |
| 22 cities in India                      | 3.16–4.14             | O\textsubscript{3} increase (+13\%); PM\textsubscript{2.5} (−43\%)                                          | More decrease in NO\textsubscript{x} compared to VOC in VOC-limited areas; more sunlight due to less PM | Sharma et al. (2020)        |
| India (Delhi, Mumbai, Chennai, Hyderabad, Bengaluru) | 3.24–4.24             | MD\textsubscript{48} O\textsubscript{3} increase in Delhi (11\%), Hyderabad (3\%), and Bengaluru (26\%); PM\textsubscript{2.5} (−10.46\%) | More decreases of NO\textsubscript{x} (compared with VOCs) reduce O\textsubscript{3} titration; enhanced HO\textsubscript{x} concentrations; increased temperature | Zhang et al. (2021)        |
| India                                   | 3.25–4.15             | Increased O\textsubscript{3} exist in most urban areas                                                                 | Reduction in the emission ratio of NO\textsubscript{x} to NMVOC; reduced night-time O\textsubscript{3} titration. | Tibrewal and Venkataraman (2022) |
| Delhi metropolitan agglomeration        | 3.25–5.17             | O\textsubscript{3} increase (+27\%);                                               | Less O\textsubscript{3} titration; higher solar radiation                                      | Dumka et al. (2021)        |
| 9 different cities in Gujarat state     | 3.24–4.20             | O\textsubscript{3} increase (+25–48\%);                                             | Less O\textsubscript{3} titration; higher insolation; warmer temperatures                       | Selvam et al. (2020)       |
| Hyderabad City                          | 3.24–4.30             | O\textsubscript{3} increase (<)                                                   | Less O\textsubscript{3} titration; the decrease in CO and NO\textsubscript{x} concentration    | Allu et al. (2021)         |
| Southern India                          | 3.25–5.3              | O\textsubscript{3} increase (<10.7\%)                                               | Less O\textsubscript{3} titration; lower fine particle loadings led to less scavenging of HO\textsubscript{2} | Kalluri et al. (2021)      |
| Delhi City                              | 3.25–4.14             | O\textsubscript{3} increases in the industrial and transport dominated locations (<10\%); PM\textsubscript{2.5} (−53.11) | Less O\textsubscript{3} titration; warmer temperatures                                        | Kumari and Toshniwal (2020) |
| Delhi, Mumbai                           | 3.25–4.15             | O\textsubscript{3} increase (<20\%);                                              | Dramatic reduction in anthropogenic activities; increase in the atmospheric boundary layer (ABL) height | Dave et al. (2021)         |
| Ahmedabad city                          | 4.10–5.1              | Non-refractory PM\textsubscript{2.5} reduction (<50\%)                             | Emission changes; increase in PBL and high wind speed                                          | Singh and Kumar (2021)     |
| New Delhi                               | 3.25–5.31             | PM\textsubscript{2.5} increase (<54.8\%)                                            | Anthropogenic pollutant sources; lockdown strictness and duration; meteorological fluctuations. | Kumari et al. (2020)       |
| Chennai, Delhi, Hyderabad, Kolkata, Mumbai | 3.25–5.11            | PM\textsubscript{2.5} increase (<19–43\%, Chennai), (41–53\%, Delhi), (26–54\%, Hyderabad), (26–36\%, Kolkata), (10–39\%, Mumbai) |                                                                                                 |                             |

\(^a\) Refers to the major lockdown period in all study periods.

\(^b\) Refers to the results during the lockdown compared to the previous years, otherwise, it is compared to the period before the COVID-19 lockdown.

\(^c\) Refers to the reasons for PM\textsubscript{2.5} concentration changes, otherwise, it refers to the reasons for O\textsubscript{3} concentration changes.
Yin et al., 2021), Baghdad, Iraq (Hashim et al., 2021), Milan, Italy (Zoran et al., 2020) and the Auvergne-Rhône-Alpes region, France (Sbai et al., 2021b). During the lockdown in Europe, O$_3$ was found to increase over more urbanized regions such as Central Northern Europe and the Po Valley (Cuesta et al., 2021; Grange et al., 2021; Matthias et al., 2021; Sbai et al., 2021b; Zoran et al., 2020;). O$_3$ photochemistry in these regions with high NOx emissions from industry and traffic was typically dominated by VOC-limited conditions and O$_3$ titration with NO was reduced when NOx emissions were reduced (Cuesta et al., 2021; Matthias et al., 2021). Numerous studies in India have also attributed O$_3$ increase to the reduction of NOx emission and less O$_3$ titration as shown in Table 2. An O$_3$ increase due to more decrease in NOx emissions compared to VOC emissions under a VOC-limited regime was also confirmed in some locations in India (Sharma et al., 2020; Tibrewal and Venkataraman, 2022; Zhang et al., 2021). It may be concluded that Kannur was most likely a VOC-limited region, as demonstrated by lockdown-induced NOx emission reduction and O$_3$ concentration enhancement. However, the validation of this hypothesis will require further investigations. A negative correlation between PM$_{2.5}$ and solar radiation was found by Chen et al. (2019), as particles can reduce the actinic flux of radiation and consequently inhibit the photolysis reactions near the surface to some degree. Thus, it was concluded that the decreased PM$_{2.5}$ concentrations during the Lock and Tri periods were conducive to higher solar radiation and helped enhance the photochemical reactions during the daytime (Dang and Liao, 2019; Li et al., 2019; Sharma et al., 2020). An O$_3$ increase during lockdown was also related to the more pronounced influence of vertical transport, which highlights the study of the associated atmospheric dynamics proposed previously by Kumar et al. (2022).

3.4. IPR analysis for fine particulate matter formation

Fig. 8 shows the hourly contributions of the individual atmospheric processes to the evolution of PM$_{2.5}$ and the hourly concentrations of PM$_{2.5}$ during the three periods. As evident in Fig. 8a, the PM$_{2.5}$ emissions and aerosol process constituted the major positive contributions to the net surface PM$_{2.5}$, accounting for a total of 48.7%, 38.4%, and 42.5% of PM$_{2.5}$ sources during the Pre1, Lock, and Tri periods, respectively (see Fig. 7). Horizontal transports during the Lock and Tri periods had a positive net effect on the PM$_{2.5}$ production, while slightly causing surface PM$_{2.5}$ export during the Pre1 periods. Surface PM$_{2.5}$ was predominantly removed by vertical transports, along with a small amount of removal by dry deposition. Photochemistry can be a significant contributor to PM formation, especially under high ozone levels, because O$_3$ can be converted to OH radicals in the atmosphere in the presence of humidity. OH radicals react with VOC and BVOC and lead to the formation of secondary aerosols that represent a significant fraction of PM$_{2.5}$ (Ortega et al., 2016; Sbai and Farida, 2019; Sbai et al., 2021a). However, the contributions of photochemistry to PM$_{2.5}$ during the three periods were not as much as expected. Photochemistry during all three periods had a net negative contribution to PM$_{2.5}$, and these negative effects became obvious with tightened restrictive measures. Considering the weak effects of cloud processes on PM$_{2.5}$, we do not discuss these two processes in this paper. It is worth noting that the PM$_{2.5}$ concentrations during both the Lock and Tri periods were lower than that during the Pre1 period, and this was primarily explained by the weaker PM$_{2.5}$ production from emission and aerosol processes. The increased PM$_{2.5}$ vertical transport rate from the surface level to the upper level was also a reason for the PM$_{2.5}$ decrease during the Lock period.

As Fig. 8b shows, from the perspective of the entire planetary boundary layer, the aerosol process, vertical transport, and emissions were the major contributors to PM$_{2.5}$. The net effects of the vertical and horizontal transports within the boundary layer were opposite to that at the surface layer, indicating a complicated distribution of transport effects in the upper layers. Therefore, the hourly PM$_{2.5}$ change rates due to the various atmospheric processes for layers 1 to 10 and the evolution of the PM$_{2.5}$ vertical profiles during the three periods are displayed in Fig. S3 to evaluate the vertical distributions. As Fig. S3 displays, the PM$_{2.5}$ emissions only existed within the first three layers, and this was related to the height of the emission source. In the first three layers, the horizontal transport and aerosol processes were the other two important sources of PM$_{2.5}$, while vertical transport was the major sink for removing the near-ground PM$_{2.5}$. Dry deposition only occurred at the first layer and serves as another sink for PM$_{2.5}$. The production rate of PM$_{2.5}$ via the aerosol process decreased as the vertical layer rose. Vertical transport contributed positively to the upper layers and negatively to the lower layers, while horizontal transport had the opposite effect. This resulted in a vertical export and a horizontal import at the surface layer. This may have been related to the local air circulation existing over Kannur. The PM$_{2.5}$ concentration had the highest value at the surface layer and decreased as the vertical layer increased for layers 1 to 6, and this could have been attributed to the contribution of primary emissions and the aerosol process.

Fig. 9 depicts the diurnal variations for the contributions of the different processes to PM$_{2.5}$ formation during the three periods. As shown in Fig. 9a, during the Lock and Tri periods, the positive contributions of emissions and the aerosol processes experienced a small increasing trend from 0700 LST to 0900 LST. However, the PM$_{2.5}$ quantities were removed due to the negative effect of horizontal transport and the negatively shifted effect of vertical transport during this period. During 0900–1200 LST, the contribution of horizontal transport transformed from negative to positive and performed as the primary contributor of the net PM$_{2.5}$, resulting in an upward trend in the PM$_{2.5}$ concentrations and a second PM$_{2.5}$ peak at 1200 LST. Later, the positive effects of horizontal transport and the aerosol process tended to weaken. Therefore, the net effects of these positive processes were not enough to balance the continuous negative effect of the vertical transport on the PM$_{2.5}$ concentrations, causing a downward trend in PM$_{2.5}$ during this period. After a period of flattening, the PM$_{2.5}$ levels rose again due to the...
positive contribution of vertical transport, emission, and aerosol processes during the nighttime. Generally, during the daytime, horizontal transport constituted the primary source of PM$_{2.5}$ on the ground, with the highest positive concentrations of 130.0 µg/m$^3$ and 70.4 µg/m$^3$ to the hourly PM$_{2.5}$ concentration during the Lock and Tri periods, respectively. PM$_{2.5}$ was primarily transported to the upper layers, especially during the daytime, with the highest removal rates of 151.6 µg/m$^3$/h and 90.5 µg/m$^3$/h during the Lock and Tri periods, respectively. Although the PM$_{2.5}$ trends during the three periods were similar, the magnitudes of the PM$_{2.5}$ changes and the contributions to the net PM$_{2.5}$ differed during the three periods. The primary difference was that the two peaks of PM$_{2.5}$ during the Pre1 period (88.5 µg/m$^3$ at 0800 LST; 60.5 µg/m$^3$ at 1200 LST) were higher than that during the other two periods. This result was primarily attributed to the higher positive effects of the aerosol process and the higher PM$_{2.5}$ emissions during the Pre1 period.

Table 2 compares the PM$_{2.5}$ reductions during the lockdown in India reported in previous studies and in the present study. The average PM$_{2.5}$ reduction (41.6%) during the Lock and Tri periods was comparable with 43.35% (averaged for Delhi, Mumbai city) reported by Kumari and Toshniwal (2020), and within the range calculated by Zhang et al. (2021) and Selvam et al. (2020). However, there were discrepancies between the results of this study and other results in Table 2 due to different study areas and the duration of the lockdown period. Previous studies in India have demonstrated the reasons for PM$_{2.5}$ reduction from the perspective of the increased atmospheric boundary layer, meteorological fluctuations, components change, and source apportionment as shown in Table 2. The changes in emissions represented a declined primary source of PM$_{2.5}$ that was most likely caused by constrained vehicular emissions and industry emissions (Du et al., 2021). Besides primary emissions, the contribution of the aerosol process is related to the secondary formation of aerosols, such as secondary nitrate and secondary sulfate formed from gaseous precursors via condensation or oxidation (Jain et al., 2020; Zhang et al., 2019). These findings are generally consistent with our findings that the decline in PM$_{2.5}$ concentration was largely caused by the weaker effects of emission and the aerosol process. In addition, the increased vertical export of PM$_{2.5}$ from the surface to the upper layer also accounted for the surface PM$_{2.5}$ decrease during the Lock period, which implied the importance of the aforementioned atmospheric dynamics reported previously by Kumar et al. (2022).

3.5. Impacts of meteorological variation and emission reduction on IPR

A sensitive simulation was conducted to assess which factor (meteorology or emission) has a more significant impact on air quality changes during the lockdown. In the sensitive simulation, the emissions were not adjusted with the scaling factors (denoted as ‘Base’ in the following description), and the meteorology was kept the same as in the Mod case. Therefore the difference between Base and Mod was due to emission changes during the lockdown. The contributions of emissions reductions were calculated by subtracting the value of Base with Mod in the corresponding period. The absolute differences between Lock and Pre1, as well as between Tri and Pre1, include contributions from both meteorology changes and emission reductions. Therefore, the contributions of meteorology in Lock and Tri days can be estimated by subtracting the emission contribution from the total absolute difference. This method was used in a previous study (Liu et al., 2020) to separate the contribution from meteorology changes and emissions reductions to PM$_{2.5}$ and O$_3$ changes during COVID-19 lockdown in China. Fig. S4 depicts the evolution of daily O$_3$ and PM$_{2.5}$ concentrations from Base, Mod, and observations. During the Pre1 period, PM$_{2.5}$ and O$_3$ were the same in Base and Mod. PM$_{2.5}$ concentrations during the Lock and Tri periods had no obvious downward trend compared with Pre1 in Base. When the emission reductions were considered in Mod, the discrepancies between the Lock, Tri, and Pre1 periods, as well as between Mod and Base were very clear. The O$_3$ difference between Base and Mod during Lock was small and more pronounced during the Tri period. As shown in Fig. S5, emission reductions could influence CHEM more significantly than meteorological variations. Since a slower consumption of O$_3$ by photochemistry was associated with surface O$_3$ increase during the Lock and Tri periods as previously described in section 3.3, it was concluded that the changes in CHEM caused by emission reduction had a large impact on O$_3$ increase during the lockdown. For PM$_{2.5}$, the decreases in PM$_{2.5}$ concentrations during the Lock and Tri periods were mainly explained by the weaker PM$_{2.5}$ production from emission and aerosol processes as previously described in section 3.4. As Fig. S6 depicts, aerosol and emission processes were primarily influenced by emission reductions rather than meteorology variations. Overall, the main causes of O$_3$ increase and PM$_{2.5}$ decrease (such as changes in photochemistry, aerosol, and emission processes) were mainly affected by emission reductions. Meanwhile, meteorological variations had important impacts on horizontal and vertical transport, especially during the Tri period.

3.6. Discussion

3.6.1. Policy implications

The constrained emission situation caused by the COVID-19 lockdown was a unique opportunity to evaluate the effects of emission reductions on air quality and to assess further air quality policies. This study utilized the scenario of lockdown-derived emission reductions to study the air quality changes in Kannur, India and utilized IPR to explore the contributions of individual physical and chemical processes. The findings provide insights into the reasons for air quality changes and have implications for local governments to formulate air pollution control measures. In Kannur, reducing anthropogenic emissions is indeed an effective way of mitigating PM$_{2.5}$ pollution. However, considering the fact that the reduction in anthropogenic emissions during the lockdown resulted in an increase in O$_3$, the local government should be cautious when formulating emission control strategies. O$_3$ mitigation strategies are highly sensitive to the local ozone formation regime that determines the type and relative amounts of precursor to be controlled. Future studies are recommended to investigate in O$_3$ formation regime and the reasonable reduction ratio of precursors to develop localized strategies for mitigating O$_3$ pollution. Since anthropogenic emission reductions can indeed alleviate PM$_{2.5}$ pollution, further research on PM$_{2.5}$ components and source apportionments
would be conducive to implementing more effective emission reduction strategies for different sources.

3.6.2. Limitations

A few limitations may affect the results of this study. The first limitation lies in the model setup and inputs. Uncertainties in the meteorology predictions could have led to some bias in the meteorological parameters, such as temperature, solar radiation, wind speed, and the boundary layer height. Higher solar radiation and temperature can promote photochemistry formation during the daytime (Chen et al., 2019; Wang et al., 2022; Yang et al., 2020; Yin et al., 2019). Wind speed and the boundary layer height are associated with the mixing degree of pollutants, and thus have great impacts on the horizontal and vertical transports of \( \text{O}_3 \) and PM\(_{2.5}\). Uncertainties in the emission inventory and grid resolution could have also led to some bias in the predictions (Hu et al., 2015; Wang et al., 2021b; Wang et al., 2019). The anthropogenic emission inventory (REAS) is subject to some uncertainties in emission factors, activity data, removal efficiencies, and the allocation of emission sources at high spatial and temporal resolutions (Kurokawa and Ohara, 2020). REAS version 3.1, with a spatial resolution of 0.25° × 0.25°, was used in this study but was still coarse for our model resolution (4 km × 4 km). Further studies are required to develop more detailed model parameters, more accurate meteorological predictions, as well as local and fine emission inventories to reduce model uncertainty and improve model performance in the future.

The second uncertainty is associated with the scaling factors (Table S1, Supporting Information) applied in this study for the different emissions species. More accurately estimating the lockdown-derived emission changes from different sources, such as vehicle emissions, industrial emissions, and residential emissions is beyond the scope of the current study. Projected emission inventories might not have represented the actual emissions accurately during the lockdown due to large uncertainties in the emission activities and the scaling factors used in this study. Future studies should identify and estimate emission changes from more detailed sources to provide quantitative information for accurate and effective emission adjustments during the lockdown.

4. Conclusions

The CMAQ model was applied to evaluate the air quality in the coastal city of Kannur, India, during the 2020 COVID-19 lockdown. Both the simulations and observations showed a decline in the PM\(_{2.5}\) concentrations and an enhancement in the \( \text{O}_3 \) concentrations during the Lock and Tri periods compared with that in the Pre1 period. The IPR analysis was employed to isolate and quantify the contributions of individual atmospheric processes to explore the causes of \( \text{O}_3 \) and PM\(_{2.5}\) concentration changes. The results revealed that the surface \( \text{O}_3 \) enhancements during the Lock and Tri periods were primarily attributable to the weaker negative effect of photochemistry, which was caused by a lesser \( \text{NO}_x \) titration effect during the nighttime and more active photochemical generation during the daytime. In contrast with previous studies, it can be concluded that the \( \text{O}_3 \) photochemistry in Kannur was in a VOC-limited condition during the study period and that \( \text{O}_3 \) titration with NO was reduced when the \( \text{NO}_x \) emissions were reduced. During the Lock and Tri periods, the surface PM\(_{2.5}\) decrease was primarily caused by the reduced emissions and the aerosol process, and this represents a reduction in the primary source of PM\(_{2.5}\) and a reduction in the secondary aerosol formation via gas-to-particle conversion. Horizontal transport and vertical transport both had a significant effect on the changes in surface \( \text{O}_3 \) and PM\(_{2.5}\). During the Lock period, the surface \( \text{O}_3 \) enhancement was also caused by the lower removal rate by horizontal transport, and the PM\(_{2.5}\) decline was also caused by the higher removal rate by vertical transport. During the Tri period, a stronger vertical import from the upper layer to the surface also accounted for the \( \text{O}_3 \) increase compared with the Pre1 period. This study demonstrates the contribution of individual atmospheric processes and estimating the causes of quality changes due to lockdown measures, and provides insights for regulatory authorities when considering the formulation of localized air pollution control policies for \( \text{O}_3 \) and PM\(_{2.5}\) in Kannur, India.

Author contributions

FY and JH designed research. FY, LH, LL, and JH conducted the simulations, DR, NT, and SK collected the data. FY, DR, and JH analyzed the data, all authors discussed the results. FY prepared the manuscript and all authors helped improve the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.119468.

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