THE ROLE OF CLIMATE AND AIR POLLUTION IN HUMAN HEALTH AND URBAN CHEMISTRY IN ASIAN CITIES

EDITED BY: Prashant Rajput, Atinderpal Singh, Jai Prakash and Manish Kumar PUBLISHED IN: Frontiers in Sustainable Cities







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THE ROLE OF CLIMATE AND AIR POLLUTION IN HUMAN HEALTH AND URBAN CHEMISTRY IN ASIAN CITIES

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Editorial: The Role of Climate and Air Pollution in Human Health and Urban Chemistry in Asian Cities

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Editorial on the Research Topic

The Role of Climate and Air Pollution in Human Health and Urban Chemistry in Asian Cities

The Asian cities are experiencing unprecedented climate, poor air quality, and human health due to existing rapid urbanization, air pollution, unsustainable land-use planning, and industrialization. Therefore, air pollution is now one of the biggest threats to human health in urban cities of Asia. The association between air pollution and human health is very complex due to atmospheric processes and the transformation of pollutants which are considered as exposure variables. Furthermore, biomass burning and climate change effects for example rise in temperature and the perturbed hydrological cycle can significantly impact the atmospheric chemistry, and human health in urban air-sheds in Asia.

A total of 10 articles are provided in this special issue, which is aimed at recent developments in the area of air pollution, urban atmospheric chemistry, and its impact on public health and climate change in Asian cities. This issue is centered on the novelty of work reported from experimental as well as modeling analysis in the field of air pollution, climate change, and human health. The manuscripts went through a rigorous, transparent, single-blinded, and interactive peer-review process involving the authors, the Reviewers, and the Guest Editors prior to acceptance for publication in this special issue. The gist of these publications is given below:

Many researchers have reported the bursting of firecrackers (FC) during the Diwali festival (celebrated on a particular date either in the month of October or November) in India which leads to further increase the air pollution above the background levels and plausibly affects the human health. During post-monsoon (October-November), the long-range transport of biomass burning emissions (LRT-BB) also affects the air quality in downwind locations in Northern India. Rajput et al. revealed that FC burst in Diwali and LRT-BB increased the daily PM_{2.5} concentration by 11 and 36%, respectively over its urban background level (286 μ g m⁻³) at Kanpur using the Lenschow-type approach on a temporal domain for the first time. Bangar et al. reported the source apportionment of PM_{2.5} from Northern India (Delhi) during the post-monsoon season and they also found that LRT-BB has the highest contribution to PM_{2.5} during this season as reported by Rajput et al. Along with LRT-BB, they identified the other major sources of PM_{2.5} such as uplifted mineral dust, vehicular emissions, road dust resuspension, secondary aerosols formation, industrial emission, coal combustion, and solid-waste burning.

The urban population is subjected to multiple exposures to air pollution and heat stress that have several negative health impacts. Indian cities are highly vulnerable to extreme weather events for example heatwaves and cold waves. A review article by Menon and Sharma highlights the use Nature-Based Solutions (NBS) to tackle the environmental issue due to their multi-functional

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Rajput P, Singh A, Prakash J and Kumar M (2022) Editorial: The Role of Climate and Air Pollution in Human Health and Urban Chemistry in Asian Cities. Front. Sustain. Cities 4:858060. doi: 10.3389/frsc.2022.858060 nature and cost-effectiveness. In their review article, they highlighted co-benefits of using NBS such as reduction in energy cost as well as conservation of biodiversity in addition to improving public health (through a reduction in air pollution and urban heat). Dutta et al. examined the impact of heat stress on the stone quarry workers. The result findings suggest that around 14% of workers were vulnerable to heat stress.

Tuladhar et al. estimated the health impacts of PM_{2.5} exposure using the WRF-Chem Model in Kathmandu Valley, Nepal. The exposure analysis indicates that 19 people could die due to lung cancer and 175 people could die due to all-cause (non-accidental) diseases due to PM2.5 exposure in December. Furthermore, their simulation estimated that reducing the 50% PM_{2.5} level in the valley could lead to a reduction in the monthly mortality by 51.4%. Pavel et al. estimated the human health risk due to the criteria pollutants in Dhaka, Bangladesh. They found that hazard quotient (HQ) values were not antagonistic (HQ < 1) while assessing acute exposure in the three age groups (infants, children, and adults). However, their study showed a significant health risk (HQ > 1) in chronic exposure for infants and children. They identified children are the worst sufferers among the age groups. Air pollution due to nanoparticles (NPs) is receiving increasing attention in scientific communities due to their strong influence on human health. Sonwani et al. provided a comprehensive review on the atmospheric NPs and their association with human health. Exposure to NPs causes the generation of ROS, resulting in cytotoxicity that leads to genotoxicity and tumorigenesis. The overproduction of ROS and the weakening of the antioxidant defense system cause oxidative stress which can trigger the release of more pro-inflammatory hormones that lead to inflammation as well as acute and chronic lung diseases.

Climate change is one of the biggest challenges of sustainability in today's world. Kaur and Pandey reviewed the present status of climate change, air pollution, and human health in Indian cities. In this review, they stated that the Indian population is experiencing adverse human health impacts due to air pollution and climate change. Further, they emphasized the role of climate change in arising extreme weather events in India. They also highlighted the use of satellite data with geospatial techniques in monitoring and mapping spatial-temporal distribution patterns of air pollution and climate change and associated health impacts. Therefore, to make sustainable cities in developing countries like India, there is a need for stringent urban planning, electric mobility, and action plans to curtail urban air pollution and improve the public health system.

The COVID-19 pandemic has affected our economic growth and health care system. Mishra et al. assessed the impact of lockdown and unlock phases on ambient atmospheric air quality parameters across 16 major cities of India covering the northto-south stretch of the country. They reported a reduction in $PM_{2.5}$ by 49% over north India during the lockdown period. Their results indicate that by adopting cleaner fuel technology and avoiding poor combustion activities across the urban cities of India a reduction in $PM_{2.5}$ up to 30% can be achieved. Another study (Yadav et al.) focused on the substantial improvement in air quality over 6 cities of the states of Rajasthan (India) during the nationwide lockdown amid COVID-19.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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Source Contribution of Firecrackers Burst vs. Long-Range Transport of Biomass Burning Emissions Over an Urban Background

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This study reports on the high-resolution data set of ground-level O_3 , surface-bound polycyclic aromatic hydrocarbons (SB-PAHs), and particle's number concentrations (range: 10 to 1,000 nm, referred to as condensation nucleus concentration: CNC) during a Diwali festival campaign (conducted from 08th to 16th Nov.2015) at Kanpur location. In this study, we have made an attempt to assess the change in atmospheric composition and chemistry (based on SB-PAHs, O₃, and CNC) during Diwali festival (11th Nov.) and compared the results with pre-Diwali (08th-10th Nov.) and post-Diwali (12th-16th Nov.) scenarios. The wind pattern and cluster analysis have revealed a quite similar feature that from 10th to 16th of November the prevailed winds were north-westerly (NW). It is noteworthy that NW-winds during post-monsoon season (Oct-Nov) favors the long-range transport of biomass burning emissions (LRT-BB) from its source region in upwind Indo-Gangetic Plain (IGP). The influence of LRT-BB emissions at the receptor site during Diwali and post-Diwali period was reflected by the substantial increase in average concentrations of PM_{2.5}, O₃ and CNC (difference has been ascertained from a two-tailed t-test). The Lenshchow-type analysis revealed that the firecrackers (FC) burst and LRT-BB emissions have lead to increase the concentrations of CNC by 54% and 86%, respectively over the urban background level. On the other hand, the FC burst and LRT-BB increased the concentrations of O_3 by 12% and 31% (over the urban background), respectively. Lenschow-type analysis revealed that FC burst and LRT-BB increased the daily PM25 concentration by 11% and 36%, respectively over its urban background level (286 μ g m⁻³). However, the SB-PAHs concentrations were found to be decreased by 6% and 2%, respectively, during the FC burst activity and LRT-BB emissions. Based on the observations pertaining to the decrease in SB-PAHs concentrations from the Lenschow-type analysis and anti-correlation between SB-PAHs and O₃ the heterogeneous-phase chemical reactivity and loss of SB-PAHs has been inferred in this study.

Keywords: PAHs, O₃, Diwali, Indo-Gangetic Plain, Air pollution, Heterogeneous-phase chemical reactivity

INTRODUCTION

Diwali festival is one the major festivals celebrated across India. It is also known as the Festival of Lights and people celebrate it with lots of enthusiasm and through cultural activities. This festival is celebrated on a particular date either in the month of October or November based on the Indian Vedic Calendar. However, worsening of air quality on Diwali festival due to bursting of firecrackers (FC) is one of the major concerns raised by many researchers (Singh et al., 2010; Joshi et al., 2016; Ambade, 2018; Izhar et al., 2018; Rastogi et al., 2019). It is worthwhile mentioning that unlike in the western and southern parts of India, the air quality in northern part of India worsens during October-November due to additional input of pollutants from the long-range transport of biomass burning emissions (LRT-BB) (Rajput et al., 2011, 2016; Kaskaoutis et al., 2014; Jethva et al., 2019; Sharma et al., 2019). Thus, LRT-BB poses a challenge in estimating the emissions due to FC burst over the urban background emission (manifested primarily by vehicular exhaust, industrial emission, and soil dust resuspension) in northern India. The overall pollution load at any given time over a city site is due to, the emissions occuring within the urban agglomeration (hereafter referred to as urban background emissions) and those getting transported from long distances (e.g., LRT-BB), atmospheric chemistry resulting into the formation of new compounds, and episodic emissions (e.g., FC burst on Diwali). To the best of our knowledge, the quantitative estimate of pollutant's concentration only due to FC, after correcting for the contribution of LRT-BB, has not been examined previously for the northern India.

Besides the characterization and impact of primary species (Menon et al., 2002; Ramana et al., 2010; Andreae and Ramanathan, 2013; Sahu et al., 2017; Satish and Rastogi, 2019), there has been a deep interest globally in looking at the features pertaining to chemical transformations occuring under ambient atmospheric conditions (Perraudin et al., 2007; Rudich et al., 2007; Kaiser et al., 2011; George et al., 2015; Nguyen et al., 2016; Pöhlker et al., 2018; Rajput et al., 2018; Rajput and Gupta, 2020). For example, a recent study assesses the secondary organic aerosols (SOA) formation due to ozone utlizing the instrumental variable analysis (IVA) (Rajput and Gupta, 2020). In fact, many studies have focussed on the atmospheric reactivity and health impacts of several types of organic compounds including the polycyclic aromatic hydrocarbons (PAHs) (Ackerman et al., 1994; Maria et al., 2004; Che et al., 2016; Ruehl et al., 2016; Singh and Gupta, 2016; Agarwal et al., 2018). The surface layer reactivity of PAHs with the atmospheric oxidants (e.g., O3 or OH radical) has been found to be asociated with an enhanced cloud condensation nuclei (CCN) activation efficiency and toxicity (of a particle) (Perraudin et al., 2007; Kaiser et al., 2011). The surface-layer oxidation reaction of PAHs plausibly leading to an enhanced CCN activation efficiency is shown through a schematic diagram (Figure 1). This study was conducted with two major objectives: (i) to quantify the contribution of FC burst vis-à-vis LRT-BB emissions to the total burden of pollutants (SB-PAHs, PM2.5, O3 and CNC) above the urban background level at central IGP (Kanpur location) and, (ii) to investigate the association between the SB-PAHs and O_3 concentrations during day and nighttime of pre-Diwali, Diwali, and post-Diwali periods.

METHODOLOGY

Field Campaign

A 9 day campaign from 08th Nov 2015 (local time: 10:00 h) to 16th Nov 2015 (local time: 06: 00 h) was conducted at Kanpur site (26.30 °N, 80.14 °E, 142 m amsl.) in central part of the Indo-Gangetic Plain (IGP). In order to assess the atmospheric chemistry and episodic emission strength from FC burst on Diwali festival day, the entire campaign has been sub-divided into three periods: pre-Diwali (08th-10th Nov.), Diwali (11th Nov.) and post-Diwali (12th-16th Nov.). In this campaign, we have measured high-resolution ($\Delta t = 1$ -min, $n \approx 10$ k data points) near ground-level O3, surface-bound polycyclic aromatic hydrocarbons (SB-PAHs), and particle's number concentrations from 10 to 1,000 nm by condensation particle counter (referred to as CNC, condensation nucleus concentration). For these measurements, three online analyzers have been utilized which were housed in the Atmospheric Particle Technology Laboratory (APTL, first floor, ~25 ft. from the ground) at the Center for Environmental Science & Engineering (CESE) building in the premises of Indian Institute of Technology Kanpur (IITK). The APTL lab remains usually maintained at temperature \sim 22°C. These instruments were kept nearby on a platform in the lab, and their inlets (separated by < 1 m) were allowed to sample air from one of the windows. The window from which the air was sampled for the real-time analysis is situated on the rear side of the CESE building and it does not faces any direct road emission. Relevant details on each instrumentation are explained below:

Real-Time Measurements of Particle's Surface-Bound PAHs

The desktop model of the photoelectric aerosol sensor (PAS 2000, **Table 1**, $\Delta t = 1$ -min, $n \approx 10$ k data points) uses a Krypton Chlorine excimer lamp that produces photons of energy 5.6 eV peaking at 222 nm wavelength (Niessner and Walendzik, 1989). These photons of 5.6 eV are utilized to photo-ionize PAHs molecules that are adsorbed onto the surface of a sampled particle (Marr et al., 2006). SB-PAHs (unsubstituted) have lower ionization energies and so get ionized upon exposure to photons from KrCl lamp whereas the gas-phase PAHs have higher ionization energies and so neither they get ionized nor be measured by PAS (photoelectric aerosol sensor) (Seki, 1989). Followed by ionization, an electric field removes the ejected electrons whereas the positively charged particles are collected on a filter element and the electric current thus generated is measured by an inbuilt electrometer. The output signal measured by the electrometer is theoretically proportional to the PAHs mass collected by the filter element. It is worthwhile mentioning here that the PAS instrument can measure only "PM surface-bound total PAHs (SB-PAHs)" and cannot provide PAH speciation which will require techniques such as gas



TABLE 1 | Specific details of the instruments deployed in this campaign.

Parameters	Instrument (Model)	Manufacturer	Flow rate (LPM)	Data monitoring/ acquisition adds-on
SB-PAHs	Real-time Desktop PAS (Photoelectric Aerosol Sensor; Model # 2000)	EcoChem Analytics	1.98	PAH DAS (Data Acquisition System, v 6.0.0)
O ₃	Ozone Analyzer (Model # 49i)	Thermo Scientific	0.65	Visual monitor and equipped with Ethernet port
CNC	CPC (Condensation particle counter; Model # 3007)	TSI Inc.	0.7	AIM (Aerosol Instrument Manager)

SB-PAHs, Surface-bound PAHs; CNC, Condensation nucleus concentration.

chromatography coupled with mass spectrometery (GC-MS) or liquid chromatography coupled with mass spectrometery (LC-MS), among others. The sample preparation followed by chemical analysis of PAHs by GC-MS or LC-MS is time-consuming and results into low-resolution data. Therefore, a sensor technique viz. PAS has been utilized for continuous monitoring of SB-PAHs in this study. The main advantages of PAS (works on aerosol photoionization technique) are its high sensitivity and ability to perform continuous (real-time) measurements with a response time of < 10 s. The PAS sensor for the detection of SB-PAHs has been developed by EcoChem Analytics, USA. The instrument was factory-calibrated for measuring PAHs concentrations up to 1,000 ng m⁻³ (limit of detection: 10 ng m⁻³) with an uncertainty of < 20%. The instrument performance and background signal check were

routinely assessed as per technical specifications provided by the manufacturer.

Real-Time Monitoring of O₃

Ozone Analyzer (Thermo Scientific; Model # 49i, **Table 1**, $\Delta t = 1$ -min, $n \approx 10$ k data points), designated by the United States Environmental Protection Agency (USEPA # EQOA-0880-047), is equipped with dual cell (sample and reference) and measures the concentration of O₃ in ambient air by UV-photometric technique. The O₃ measurement technique is a well-established technique, the details of which can be found in several papers, e.g., Lal et al. (2000). Briefly, O₃ molecule absorbs UV photon at a wavelength of 254 nm and the quantum of absorbed UV photons is directly proportional to the O₃ concentration. The instrument has a response time of 20 s with a detection limit of 5 ppb and can



Variable	Pre-I	Diwali	Div	wali	Post-Diwali	Diwali
	Day-Value	Night-Value	Day-Value	Night-Value	Day-Value	Night-Value
¹ RH (%)	57 ± 15	82 ± 4	56 ± 15	78 ± 4	66 ± 11	79 ± 5
¹T (°C)	26 ± 4	19 ± 0.5	26 ± 3.8	19 ± 0.5	22 ± 3.6	17 ± 2
¹ Wind (m/s)	5.0 ± 3.0	0.5 ± 0.4	11.0 ± 6.3	3.2 ± 1.6	3.9 ± 3.8	0.4 ± 0.3
² BLH (m)	$1,819 \pm 671$	$1,023 \pm 458$	$1,811 \pm 683$	965 ± 589	$1,373 \pm 221$	834 ± 364
² Sol flux (W/m ²)	536 ± 56	N/A	545 + 61	N/A	560 ± 41	N/A

TABLE 2 | Summary of meteorological conditions (Avg. \pm SD) during the period of measurements.

¹ Data retrieved from on-campus (@IIT Kanpur) weather station; ² Data retrieved from National Oceanic and Atmospheric Administration (NOAA). BLH represents boundary layer height and Sol flux is solar flux.

measure up to 200 ppm. The averaging time for data retrieval was set to 1 min (= 60 s). At the inlet, a Teflon particulate filter was mounted to not allow the entry of any particle in the gas analyzer. The measuring principle of O_3 analyzer is based on the Beer-Lambert law. The calibration and zero checks were performed as per the manufacturer's specifications for data quality control and assurance. The uncertainty on O_3 measurement is < 5%.

Real-Time Monitoring of CNC

Condensation particle counter (CPC model 3007, TSI Inc., **Table 1**, $\Delta t = 1$ -min, $n \approx 10$ k data points) is a portable condensation nucleus counter (CNC) which measures the condensation nucleus concentration (CNC) in the size range of $0.01-1\,\mu$ m. The field performance and single particle detection efficiency of this instrument are provided elsewhere (Hämeri et al., 2002; Devi et al., 2013). This instrument was factory calibrated and is capable of measuring particles concentrations as high as 10^5 cm⁻³. The zero counts were routinely checked by connecting the CPC monitoring unit inlet with a High-Efficiency Particulate Air (HEPA) filter and we were always satisfied to conduct the measurements. The TSI CPC-3007 is operated with isopropyl alcohol as a condensing fluid. The uncertainty of cumulative CNC measurements by CPC was < 1%.

Lenschow-Type Analysis

The Lenschow-type analysis was carried out for each parameter viz. O₃, CNC, PM_{2.5}, and SB-PAHs. The details of PM_{2.5} data set can be found elsewhere (Rajput and Gupta, 2020). Briefly, the hourly averaged data set was segregated into 3-different bins corresponding to UB (urban background), UB+FC+BB (urban background+firecrackers burst+biomass burning contribution), and UB+BB (urban background+biomass burning contribution) depending on their association with wind profile and information on FC burst (on Diwali). The north-westerly wind system favoring LRT-BB emissions was marked for the BB contribution. The UB was represented by the data set which has neither any contributions from FC burst nor from the LRT-BB. For details on finger-printing of sources, based on the analysis of air pollutants data coupled to prevailing winds, the reference is made to the original work by Lenschow et al. (2001).

RESULTS AND DISCUSSION

Wind Pattern Analyses

Real-time diurnal measurements have encouraged us to look into more detailed features of atmospheric composition variability during the campaign. The wind direction and wind-speed (refer to scale at the bottom, **Figure 2**) along with the statistics (given on each panel) were studied through the wind-rose plot. Statistical and data analysis have been carried out utilizing openair package in R software (Carslaw and Ropkins, 2012). The frequency pattern of wind along with its direction and speed are shown here for entire sampling dates during daylights (aka daytime: from 07:00 h to 17:00 h local time, top panel) and nighttime (from 00:00 h to 06:59 h and from 17:01 h to 23:59 h; bottom panel). The winds for each one of the sampling dates (i.e., from 08th to 16th Nov) are shown under the dedicated panel both for the daylight and nighttime periods (**Figure 2**).

During the entire campaign, the winds were calm for most of the nighttime hours (80-93%) with an average speed < 1 m s⁻¹, exception being on Diwali festival night (average speed = 3.2 m s^{-1}). The wind patterns during daylight hours suggested that on 08th and 09th Nov (pre-Diwali) winds were easterly to north-easterly whereas on the 10th Nov (also a pre-Diwali period) the wind-direction changed to north-to-northwesterly. During the entire pre-Diwali period (08th-10th Nov) the daytime wind speed was $< 10 \text{ m s}^{-1}$. More interestingly, the winds on Diwali festival day (11th Nov) continued blowing from the north-west direction with some contributions from south direction too. As far as the wind speed is concerned, it was > 15 m s⁻¹ with an average value of 11 m s^{-1} on the 11th Nov (Diwali festival daylight hours). During rest of the dates (post-Diwali from 12th to 16th Nov) the prevailed winds were mainly north-westerly both during daylight and nighttime hours (Figure 2). It is worthwhile mentioning that during October-November period the north-westerly winds favor transport of emissions from source-region of open biomass (paddy-residue) burning, active in upwind IGP, to the downind locations and marine atmospheric boundary layer over the Bay of Bengal (Kaskaoutis et al., 2014; Rajput et al., 2014). Furthermore, for the entire campaign, during daylight hours the winds were calm for < 22% of the time, exception being on 10th Nov (calm wind: 33%). A day-night summary of meteorological conditions



FIGURE 3 | Air-mass cluster analysis and fire-count imageries (shown by red-circles, over source region of biomass burning) during (A) pre-Diwali, (B) Diwali, and (C) post-Diwali periods.



FIGURE 4 | Diurnal profile of simultaneously measured surface-bound PAHs, ozone, and CPC-based particle's number concentration (CNC) during entire campaign.

during pre-Diwali, Diwali, and post-Diwali periods are given in Table 2.

Fire-Count Imageries (Over the Source-Region of Biomass Burning Emissions in IGP) and Air-Mass Cluster Analysis

MODIS (Moderate Resolution Imaging Spectroradiometer sensor, onboard NASA Terra and Aqua satellite) fire-count imageries (spatial resolution: 10 km) showing an intense open biomass (agricultural-waste paddy-residue) burning activity in north-western part of during the study period are shown in Figure 3. This observation is quite consistent with earlier observations reporting massive emissions of air pollutants from agricultural-waste burning in upwind IGP (Rajput et al., 2014, 2018). The details on land-use activity pattern and mapping of source-region of biomass burning emissions in upwind IGP are adopted from previous studies and shown in the Supplementary Material. Furthermore, a GIS-based open-source software viz. TrajStat was utilized for the air-mass cluster analysis (Figure 3). The cluster analysis is a widely used tool to understand the influence of cluster of trajectories on ambient level of air pollutants at the receptor site (Bansal et al., 2019; Rajput and Gupta, 2020). Briefly, in the cluster analysis method, the measured air pollutant concentrations are assigned to the corresponding trajectories and the nearest trajectories are clustered according to an angle distance function (Sirois and Bottenheim, 1995). Relevant details on cluster analysis are given elsewhere (Wang et al., 2009). To carry out the cluster analysis, a 5-day air-mass back trajectories (AMBTs, GDAS 0.5° m, @ 1,000 m above ground level) data have been utilized. These AMBTs were retrieved from NOAA HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Rolph, 2003; Stein et al., 2015).

One of the observations from **Figure 3** relates that if a trajectory cluster is passing more frequently through NW-direction to the receptor site then it is expected to have elevated levels of air pollutants from LRT-BB as compared to the case when trajectories were arriving from other directions. Let us now analyze the clusters shown in **Figure 3**. During Pre-Diwali period (**Figure 3A**), there was very little influence of NW-air masses (11%) but more importantly not passing through the source-region of biomass burning emission. It is important to mention here that these 11% NW-air masses were prevailed on 10th Nov. Furthermore, during the Post-Diwali period (**Figure 3C**), majority of air masses (81%) were originating from source-region of biomass burning emission and arriving from NW-direction



pre-Diwali, Diwali, and post-Diwali periods.

Source C	Contribution	Over	Urban	Agglomeration
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TABLE 3 Concentration [Avg. \pm SD; median; range (no. of samples)] of O ₃ ,
SB-PAHs and CNC during the three events (pre-Diwali, Diwali, and post-Diwali).

Events	O ₃ (ppb)	SB-PAHs (ng m ⁻³)	CNC (cm ⁻³)
pre-Diwali	26.5 ± 20.5; 17.5; 0.0–77.3 (n = 3,652)	45.5 ± 25.1; 44.7; 9.6–134.1 (<i>n</i> = 3,666)	12,069 ± 1,499; 11,880; 9,088–16,652 (n = 3,126)
Diwali	33.9 ± 22.3; 29.0; 6.7–91.0 (n = 1,380)	43.7 ± 21.3; 44.1; 5.9–80.7 (n = 1,396)	15,689 ± 2,899; 14,454; 10,406–23,628 (n = 1,240)
post-Diwali	37.3 ± 16.0; 37.1; 0.7–134.1 (<i>n</i> = 5,919)	41.0 ± 12.0; 34.1; 0.0–104.3 (n = 5,767)	22,189 ± 7,831; 19,844; 9,088–65,410 (<i>n</i> = 5,078)

to the receptor site. Summing up, clubbing the data set from our campaign, satellite observations on fire-counts along with the cluster analysis, it can be summarized that the receptor site (at Kanpur) was influenced with the LRT-BB emissions through NW-direction (from upwind IGP) mainly during post-Diwali and Diwali periods.

Co-variability Analysis of SB-PAHs, $O_{3,}$ and CNC

The co-variability features of SB-PAHs (n = 10,829), O₃ (n =10,951), and the sub-micron particles number concentrations (CNC, n = 9,444) are shown in Figure 4. It is evident from this figure that the high abundance of CNC (i.e., submicron particles, orange dots) was associated with higher concentration of O₃ (blue dots) and with the lower concentration of SB-PAHs (black dots, Figure 4). Thus, field based observations revealed that peaks in O3 concentration were associated with dips in SB-PAHs concentration. This observation is much more pronounced during Diwali and post-Diwali period (i.e., from 11th to 16th Nov). Further features in this regard have been discussed in the following section. The summary of the data set is shown in Figure 5 and also given in Table 3. We have carried out statistical two-tailed *t*-test analysis to infer about difference in measured species concentrations during Diwali as compared to those in Pre-Diwali and Post-Diwali periods. Accordingly, comparing the concentrations during Diwali vs. Pre-Diwali it revealed that O₃ (t = 11.1; p < 0.0001), SB-PAHs (t = 2.4; p < 0.05), and CNC (t =53.9; p < 0.0001) were significantly different during these periods. Furthermore, comparing the concentrations during Diwali vs. Post-Diwali periods it revealed that O_3 (t = 4.5; p < 0.0001), SB-PAHs (t = 6.3; p < 0.05), and CNC (t = 28.7; p < 0.0001) were also significantly different during these periods.

Correlation Analyses of O_3 vs. SB-PAHs as a Function of CNC

A 3-D linear correlation plot is shown in Figure 6 for three events (pre-Diwali, Diwali, and post-Diwali) and two periods

(daylight and nighttime). The first assessment on maximum concentrations revealed that O_3 was < 150 ppb, SB-PAHs is $< 150 \text{ ng m}^{-3}$ and CNC is $< 66 \text{ k cm}^{-3}$ during the entire campaign (Figure 6). The CNC was $< 20 \text{ k cm}^{-3}$ during pre-Diwali, $< 30 \text{ k} \text{ cm}^{-3}$ during Diwali and $< 66 \text{ k} \text{ cm}^{-3}$ during post-Diwali (CNC concentration pattern: post-Diwali >Diwali >pre-Diwali). Furthermore, the maximum concentrations of CNC were observed in nighttime hours during the post-Diwali period (12th-16th Nov). The correlation analyses revealed that during daylight hours, O3 and SB-PAHs were significantly anticorrelated on pre-Diwali ($R^2 = 0.57$, p < 0.05), Diwali ($R^2 =$ 0.71, p < 0.05) and post-Diwali period ($R^2 = 0.71$, p < 0.05). During nighttime hours, the anti-correlation was found to be weak during pre-Diwali ($R^2 = 0.09$, p > 0.05) and post-Diwali period ($R^2 = 0.37$, p > 0.05). However, the anti-correlation was found to be strong during Diwali ($R^2 = 0.71$, p < 0.05). It is worthwhile mentioning here that a previous study has suggested the production of ozone from firecrackers burst (Attri et al., 2001). Summing up, the linear correlation analysis revealed that SB-PAHs and O_3 have moderate-to-high association (p < p0.05) during all the period exception being that on pre-Diwali and post-Diwali nighttime while the anti-correlation was weak (p > 0.05). Furthermore, the CNC concentrations showed a large gradient in the correlation plot during Diwali and post-Diwali (varying from $\sim 10 \text{ k}$ to 65 k particles cm⁻³) as O₃ increases and SB-PAHs decreases. However, the overall variability in CNC during the pre-Diwali period, varying from ~10k to 16 k cm⁻³, is guite less as compared to those in Diwali and post-Diwali periods (also refer to Figure 5). It is important to note here that maximum CNC concentrations were observed during post-Diwali followed by Diwali and then pre-Diwali. Toward this, the explanation for higher abundance of CNC during Diwali period is attributable to additional input (i.e., besides urban background emissions) from FC burst and LRT-BB emissions. However, the higher CNC during post-Diwali period was by-and-large due to additional input from LRT-BB. It is worthwhile mentioning that the poor ventilation and shallower boundary layer height could also be responsible for a small fractional rise in the air pollutant's levels during the post-Diwali period (Table 2). The anti-correlation between SB-PAHs and O₃ could be attributable to chemical oxidation of SB-PAHs by O₃. It has been suggested previously that such chemical reactions have implications to enhanced CCN activity and toxicity of ambient aerosols (Perraudin et al., 2007; Kaiser et al., 2011).

Lenschow-Type Analysis: Assessment of Predominant Source Impact Over an Urban Background

Before we discuss on the Lenschow-type analysis and its application for quantifying emissions impact above the urban background level, let us first understand the need for applying this analysis. One of the simple ways to do that is to look into similar type of studies (e.g., from previous Diwali campaigns) and their conclusions drawn. There are two things which we need to reiterate here that (i) Diwali is celebrated during a day in October–November period and, (ii) almost the entire IGP experiences massive emisssions throughout these 2 months period from LRT-BB. Now, let us discuss briefly how the previous studies around Diwali period have assessed the change in pollutants concentration due to the FC burst. Previous researchers have documented very systematically the ambient concentrations of air pollutants [both particulate matter (PM) as well as trace gases] around Diwali period (Singh et al., 2010; Chatterjee et al., 2013; Ambade, 2018). For example, a study from an upwind IGP location at Delhi has documented pollutants concentrations for 6 years from 2002 to 2007 (Singh et al., 2010). Another study from a downwind location at Jamshedpur has documented PM and trace gases concentrations around Diwali period in 2014 (Ambade, 2018). From a further downwind location at Kolkata in IGP, a study has reported lots of species including metals, ionic composition in PM_{10} and SO_2 around Diwali period



FIGURE 6 Linear regression analyses (p < 0.05, exception being on pre-Diwali and post-Diwali nighttime when p > 0.05) of O₃ (X-axis, in ppb) vs. SB-PAHs (Y-axis, in ng m⁻³) by levels of CPC-based particle's number concentration (CNC, Z-axis in cm⁻³ shown on a scale at the right side of the plot) during pre-Diwali, Diwali, and post-Diwali periods for both daylight and nighttime hours.

TABLE 4 Lenschow-type analysis to estimate episodic source impact over the local background emis	nission in central IGP at Kanpur.
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Sr. No	Event/Analysis	Emission	CNC (cm ⁻³)	O ₃ (ppb)	SB-PAHs (ng m ⁻³)	^{\$} PM _{2.5} (μg m ⁻³)
1	Pr-D*	UB (no FC + no BB)	$11,915 \pm 396$	28.4 ± 7.2	41.9 ± 4.8	286 ± 74
2	D	UB + FC + BB	$15,689 \pm 2,899$	33.9 ± 22.3	43.7 ± 21.3	359 ± 71
3	Po-D	UB + BB	$22,189 \pm 7,831$	37.3 ± 16.0	41.0 ± 12.0	390 ± 172
4	(Po-D-Pr-D)/Pr-D	BB/UB	0.86	0.31	-0.02	0.36
5	(Po-D–D)/Pr-D	FC/UB	0.54	0.12	-0.06	0.11

*In view of NW winds on 10th Nov, for Lenschow-type analysis the data set measured only on 08th and 09th Nov are included to represent the impact of urban background (UB) emissions. FC, firecrackers burst; BB, biomass (paddy-residue) burning emission through long-range transport (LRT-BB); D, Diwali; Pr-D, pre-Diwali; Po-D, post-Diwali. *For details on PM_{2.5} data set, the reference is made to our recent publication (Rajput and Gupta, 2020).

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in 2010 (Chatterjee et al., 2013). It is important to mention here that all these aforementioned studies have compared on a 1-to-1 basis the pre-/post-Diwali emissions with Diwali emissions and attributed the change in a particular polutant's concentration on Diwali due to the FC burst. These studies from various locations in IGP concluded that due to FC burst the pollutants concentrations do rise by a factor of 1.5– 3 or 50–300%. However, if these previous studies (from the IGP) would have accounted for massive emissions from LRT-BB then the actual rise in pollutants concentrations due to the FC burst could have been quantified. With this regional background information, here we present an approach called the Lenschow-type analysis which would serve the purpose of quantifying emissions due to FC burst and LRT-BB over the regional background.

Lenschow et al. (2001) proposed an approach in 2001 to quantitatively estimate the contribution of PM sources spiking pollutants concentration above the regional background level. Their idea was basically to conduct a set of ambient PM measurements (spatially resolved) representative of regional background and those capturing records for episodic or longrange transported source components. Subsequently, grouping the data set into bins of sources corresponding to regional background emission and then estimating the contribution of episodic source and long-range transport was suggested in their study. The Lenschow-type analysis (Table 4) has been carried out to estimate quantitatively the impact of additional sources leading to increase (above the UB) the levels of air pollutants (Lenschow et al., 2001). It is worthwhile mentioning here that in this study we have applied the similar approach but on a temporally resolved ambient data set. Based on the Lenshchowtype analysis, we have estimated that only due to FC burst the CNC concentrations were increased by 54% whereas due to the impact from LRT-BB the CNC concentrations were increased by 86% over the urban background concentration level (at the receptor site). Thus, LRT-BB emissions appeared to be the major source (as evident from the increase in CNC concentrations by 86% than the urban background) of air pollution during the study period and results in enhancing the CNC burden by a facor of 1.6 when compared to that with the FC burst. Likewise, Lenschow-type analysis further revealed that O3 concentrations increased by 31% and 12% due to the LRT-BB and FC burst, respectively, over the urban background emission. However, in sharp contrast the SB-PAHs concentrations were found to be decreased by 2 and 6% (over the urban backgound emission), respectively, during the LRT-BB emission and FC burst (Table 4). Lenschow-type analysis also revealed that the urban background concentration of daily PM2.5 was 286 µg m^{-3} (Table 4). The FC burst and LRT-BB increased the daily PM_{2.5} concentration by 11% and 36%, respectively over its urban background level (286 μ g m⁻³) (Figure 7). The application of Lenschow-type analysis in this study from IGP has led to the new insights of estimating the impact of additional sources spiking the pollutants concentrations over typical emissions within an urban agglomeration.

CONCLUSIONS

In this study, we have conducted a 9 day (from 8th to 16th Nov 2015) long real-time (high-resolution: 1 min, $n \approx$ 10k data points) measurement campaign assessing the diurnal profiles of SB-PAHs, O3 and CNC during pre-Diwali (8th-10th Nov), Diwali (11th Nov) and post-Diwali periods (12th-16th Nov). Wind analysis indicated that initially on 08th and 09th Nov the winds were easterly-to-north-easterly with intensity (mostly $< 5 \text{ m s}^{-1}$) whereas from 10th onwards the winds were predominantly north-westerly (varying from 10 m s^{-1} to > 15 m s⁻¹). A quite similar inference has been made based on the air-mass cluster analysis. The source-region of large-scale post-harvest agricultural-waste burning emissions lies in the north-west direction of the study region. Under prevailing NW winds, the LRT-BB emissions appeared to change the atmospheric composition and chemistry over the central IGP. Higher concentrations of CNC, PM2.5, and O3 during Diwali are attributable to the additional input from FC burst, and substantial contribution from LRT-BB emissions. However, higher concentrations of CNC, PM2.5, and O3 on post-Diwali period are mainly due to the LRT-BB emissions.

The daily average concentrations of O_3 and CNC exhibited a quite similar pattern: post-Diwali >Diwali >pre-Diwali, whereas the SB-PAHs showed a different pattern with the highest concentration during pre-Diwali >Diwali >post-Diwali (trends are confirmed based on the two tailed *t*-test). Their diurnal profile also relates to the similar findings that when CNC and O_3 concentrations peaked up then the SB-PAHs showed a dip and vice-versa. The daylight and nighttime linear correlation analyses of SB-PAHs and O_3 as a function of CNC were carried out, and the results showed a moderate to strong anticorrelation during the entire study period, with exceptions being observed for pre-Diwali and post-Diwali nighttime hours

when the correlation was weak. Based on the Lenshchow-type analysis, it has been estimated from this study that the CNC and O₃ concentrations were increased (over UB) by 86 and 31%, respectively due to the LRT-BB emissions. Furthermore, only due to the FC burst the CNC and O₃ concentrations were increased by 54 and 12%, respectively. Lenschow-type analysis further revealed that FC burst and LRT-BB increased the daily PM2.5 concentration over the urban agglomeration by 11% and 36%, respectively. However, the SB-PAHs concentrations were found to be decreased by 2 and 6% during LRT-BB emissions and FC burst, respectively. This study, highlighting the plausibility of surface-layer (heterogeneous-phase) reactivity of SB-PAHs with O3 has potential implications to enhanced particle's toxicity and CCN activity of aerosols over the IGP. Future studies would be required to examine the causal inference of the chemical reactivity of individual PAH.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

PR and TG conceptualized this study. The data analysis presented in this paper was performed by PR. The manuscript has been

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written by PR. All authors were involved in collection of data during entire campaign used in this analysis. All authors discussed the results and commented on the manuscript.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/frsc.2020. 622050/full#supplementary-material

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Perceived Thermal Response of Stone Quarry Workers in Hot Environment

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Dutta P, Chorsiya V and Nag PK (2021) Perceived Thermal Response of Stone Quarry Workers in Hot Environment. Front. Sustain. Cities 3:640426. doi: 10.3389/frsc.2021.640426 **Introduction:** Impact of heat on health of workers goes unrecognized by the virtue of the indispensable fact that every individual has varied perception and tolerance capacity. The present study determine the physiological signs with perceived subjective responses under the thermal stress.

Materials and Methods: The study was spread on open field stone quarry workers (N = 934) during the summer (May to June), post monsoon (September to October), and winter (December to January).

Results: In the summer months, dry bulb temperature range from 36.1 to 43.2°C and the distribution of Wet Bulb Globe temperature (WBGT) outdoor values were outlier-prone than normal distribution indicated heat vulnerability. The environmental effect on weighted average skin temperature (T_{sk}) local segmental T_{sk} and deep body temperature (T_{cr}) were greater than the effects that might be attributed to work severity. The tolerance time level in summer months (65 ± 13 min at WBGT $35 \pm 2.3^{\circ}$ C) was less than in other two season. About 85% of workers in summer, 68% in post monsoon and 79% in winter recorded working heart rate greater than 90 beats/min. Physiological and subjective responses to heat stress indicated that during summer month the workers complained of excessive sweating (93.5%), feeling of thirst/dry mouth (88.7%), elevated Core temperature (T_{cr}) (58.7%) and decreased working capacity (75.6%). The observation found that around 14% workers were vulnerable to heat stress and the workers had no knowledge to mitigate the heat related illnesses.

Discussion and Conclusions: The stone quarry work as compared to other outdoor workers have environmental adversaries which becomes confounding variables in the study of such occupations. There was significant difference (p < 0.001) as far as the physiological and thermoregulatory responses were concerned in three different months of investigation.

Keywords: stone quarry, heat wave, WBGT, tolerance time, perceived response

INTRODUCTION

Heat waves are becoming increasingly severe and frequent, exacerbated by climate change threatening health and livelihood directly or indirectly (Dutta and Chorsiya, 2013; Azhar et al., 2014; Nag et al., 2014). Over a million workers are employed in quarrying and related activities in India (Saiyed and Tiwari, 2004). Types of rock extracted from quarries include cinder, chalk, china clay, clay, coal, coquina, construction aggregate (sand and gravel), globigerina limestone (Malta), granite, grit stone, gypsum, limestone, marble, phosphate rock, and sandstone. A number of sand stone quarries are located in different states of India, e.g., Rajasthan, Madhya Pradesh, Gujarat, Orissa, Karnataka, Tamil Nadu, and Andaman and Nicobar islands. Sand stone quarry is an open excavation from which the stone is obtained, by labor-intensive and strenuous methods. The workers use heavy hand tools for extracting process (layers of hard rocks) and perform many manual material handling tasks like breaking, drilling the hard rocks and lifting/carrying those for loading and unloading to transport to desired destination. The workers are exposed directly under the sun throughout their working day. Huge hammers or mechanical drilling are used to separate the stone blocks. Grecchi et al. (2009) have found that the traditional working methods cause musculoskeletal pain and discomfort among the stone quarry workers due to awkward postures and lifting of heavy weights. Epidemiological surveys on stone cutters and carvers found that the hand arm vibration induced white finger, sensor-neural, and musculoskeletal symptoms among the workers (Griffin et al., 2003; Makoto et al., 2005; TaMrin et al., 2012). Mathur (2005) reported that the average life span of stone quarry workers is \sim 10 years less than their fellow villagers who never worked in quarries. In the western part of India summer temperatures in stone quarries often exceed 45°C, indicating the risks of heatinduced illnesses and disorders among workers, with the relative vulnerability of young and elderly workers.

Literature reviewed suggested enough evidence to demonstrate the increasing certainty that climate change significantly aggregates the probability of extreme weather conditions, most often in directions that lead to dangerous health consequences especially to people who carry out heavy physical labor as a part of their daily jobs like steel plant, power plant, forge plant, etc. (Krishnamurthy et al., 2017; Varghese et al., 2018). In developing countries like India these workers are generally migratory and work on daily wages. When the ambient temperature exceed that of body temperature (37°C), the body loose heat by evaporation or sweating by the mechanism of thermoregulation controlled by the hypothalamus section of the human brain But, humidity affects this thermodynamic stability by limiting sweat evaporation and heat loss, which creates health impacts and loss of work capacity among the exposed workers (Saghiv and Sagiv, 2020). A thermodynamic model of heat balance is applied, with due account of heat exchanges through the segmental and compartmental interfaces of the human body, microclimatic and outer environment (Nag et al., 2007).

During the hot season, because of the strenuous physical activity and climatic condition the workers in stone quarry

accumulate heat load and heat stress which effects their occupational health and work capacity. The health hazards get severe if the person is exposed higher temperature for a longer time. With increasing temperature in future it is likely that the health of vulnerable occupational groups like stone quarry workers are big challenge. Studies have found that the work environment higher than temperature 40°C causes human discomfort and high mortality (Steeneveld et al., 2011; Petitti et al., 2015; Bunker et al., 2016). Literature related to response of extreme climate exposure of workers in stone quarry is lacking. Since the combined load of strenuous physical work and exposure to extremely hot environment have negative impacts on human health and safety, the present study focused on generating epidemiological data on the heatexposed stone quarry working population, with reference of biophysical perspective. Furthermore, these findings are needed to be substantiated by the subjective symptoms reported as perceived response under the thermal stress which underlies the utility of the present research work.

MATERIALS AND METHODS

Ethics

The written informed consent to participate in the study was taken as per the Indian Council of Medical Research (2000) ethical guidelines from the individuals (as all were above 18 years) for the publication of any potentially identifiable images or data included in this article.

A total of 934 men in the age range between 18 and 60 years were selected in the present crosssectional study of stone quarry workers from western part of India (Figure 1). Environmental and health risk surveillance were undertaken in stone quarry works, during the months of summer (May to June, N =521), post monsoon (September to October, N = 214), and winter (December to January, N = 199) months. Workers underwent seasonal extremes of climate scenarios and experience hardships which may sometimes fall beyond their coping levels, resulting in heat injuries. Direct measurements of the thermometric parameter include ambient dry bulb temperature (T_a), wet bulb temperature (T_{wb}), dew point, wind velocity and globe temperature (Tg) were measured by Wet-Bulb Globe Temperature (WBGT) Monitor, Delta OHM (HD 32.1, Thermal Microclimate, Italy) and Relative Humidity/Temperature data by Lascar EL-USB-2-LCD, Sweden for several hours of observation period and continued for a number of days at each workplace. The environmental warmth was expressed in terms of WBGT index (Liljegren et al., 2008). The locations of stone quarry were same in the summer, post monsoon, and winter seasons during the investigations. However, same workers could not be followed up in different seasons since the workers were migratory and casual laborers worked on daily wages.

In order to ascertain susceptibility of workers to heat stress, a checklist enquiry was introduced for health risk surveillance, heat exposure- related morbidity of the work groups, including environmental warmth assessment, physical fatigue and perceived effort. The workers' subjective responses



FIGURE 1 | Stone quarrying activities in the open cast mine: (A) carrying stone slab; (B) Removing slab for breaking; (C) self-made stone shelter in the mine; (D) Breaking stone with hammer; (E) Manually lifting slab for breaking; (F) Loading stone slab; (G) Drilling stone; (H) Separating stone slabs.

were recorded on a five-point Likert scale, and a score of 4 and 5 were taken as an indication of high strain response. Every worker was subjected to physiological variables measurement that included heart rate responses, blood pressure measurements, thermographic profile of the skin areas (T_{sk}) and deep body temperature (T_{cr}). The thermographic profile of the skin (T_{sk}) areas were recorded, using ThermoCAM, FLIR system (Sweden) from four exposed sites, i.e., head, hand, trunk, leg, and back trunk. The measurements were repeated thrice, i.e., pre-exposure and at an interval of about 2-3h during work. Heart rate was measured by polar heart rate meter (S810TM Polar Electro Oy, Finland); deep body temperature (T_{cr}) as oral temperature by thermometer and OMRON digital BP instrument was used to record the blood pressure during the work. The polar heart rate monitor and cuff of BP apparatus was tied to the chest and the arm of the worker respectively before the commencement of the work. The polar heart rate automatically record the heart rate and when BP need to monitor the OMRON BP apparatus was connected to the lead of the cuff and BP was measured in less than a minute for a single worker without interrupting their main work process. The data of Polar heart rate meter was later transferred to the computer. During the occupational exposures, the workers wore light clothing-wearing shorts, trouser or a lungi/dhuti (a loose fabric wrapped around join at ankle length) and a half-sleeve banian or t-shirt, with clothing insulation value approx. 0.6 clo (Summer clothing insulation unit).

The algorithm allowed computation of heat exchange parameters, including heat conductance, metabolic load, effective heat load, body heat storage, and the overall rate of change of body core temperatures. These dimensions led to the prediction of the limits of tolerance to work in hot environments. This algorithm is based on premise of biophysical approach propagated by Nag et al. (2007), utilizing the heat exchanges through different avenues across the segment (i.e., head, trunk, arm, hand, leg and feet) and body layers- blood, core (viscera plus skeleton), muscle, fat and skin (i.e., 6 segment × 5 layers = 30 compartments) (Nag et al., 2007). These are the following equations with thermodynamic model of heat balance of each segment (where storage δ H = 0) adopted from Nag et al. (2007).

$$\begin{split} &Y\Delta T/\Delta t = (V\rho \times S)_{Blood} \times (T_{Blood}) + \Delta M \\ &- [\{K_{Blood-core}(T_{Blood} - T_{core}) + K_{Core-Muscle} \\ &(T_{Core} - T_{Muscle}) + K_{Muscle-Fat}(T_{Muscle-}T_{Fat}) \\ &+ K_{Fat-Skin}(T_{Fat} - T_{Skin}) + H(i)(T_{Skin-}T_{Environment})\} \\ &\times SA + (C_{Res} + E_{Res} + E_{Skin})] \end{split}$$

Where Y, product of compartmental mass and specific heat, $\Delta T/\Delta t$, change in temperature with time, V, volume (liter), ρ , density (kg/L), S, Specific heat of blood (W h/kg °C), ΔM , (total-basal metabolic energy, W.h), K, conductance of body compartments (W/m².°C), T, resultant body temperature (°C), H(i), combined heat transfer coefficients of segments (W/m².°C), SA, surface area (m²), C_{Res} and E_{Res}, resoiratory heat loss through convention and evaporation (W.h), E_{skin}, evaporation heat loss for skin (W.h).

STATISTICAL ANALYSIS

Data analysis was performed using SPSS statistical software, version 16.0. Descriptive statistics was applied to understand the charactersitics of temperature profile and physiological variables of the workers. One way ANOVA was used to study the differences of these variables among three different seasons. Percent prevalence was calculated for perceived sign and symptoms responses of the workers. The level of significance was set at p < 0.05.

RESULT

The open-field day-time ambient conditions are presented in **Table 1**. The percentile distribution, skewness, and kurtosis values of WBGT estimated during the three seasons of investigation reflected the variations in environmental warmth. During summer (May to June), the distribution of WBGT outdoor values was more outlier-prone than the normal distribution. The positive kurtosis indicated a relatively peaked distribution. The WBGT values spread out more to the right from the proximity of the mean ($35 \pm 2.3^{\circ}$ C) and thereby indicating a component of heat vulnerability of the sample population concerned. The environmental data were found to be statistically different in three seasons (summer, post monsoon, and winter) of the investigation. The workers had mean body weight 53.8 \pm 9 kg and body surface area 1.6 \pm 0.1 sqm.

Beside variation in the environmental conditions, the magnitude of physiological responses of the stone workers attributed to the combined stress of environmental exposure and the intensity of the work performed, with the potential to health consequences. A comparison of local T_{sk} and weighted average T_{sk} of workers in summer, post monsoon and winter are in Table 2, indicating their T_{sk} responses differed significantly. During the summer and post-monsoon months, the 5th-95th percentile values of T_{sk} of varied from 30.1 to 40°C and 32.7 to 37.3°C, respectively. Whereas, in the winter months, the 5th–95th percentile values of T_{sk} of local areas ranged from 24.6 to 34.7°C. The profile of segmental T_{sk} indicated the relative space for adjustment against the extent of the core temperature buildup of the workers. Table 2 also includes the weighted average T_{sk} of the whole body, which was obtained from the surface area and sensitivity weighting of local area T_{sk}. The weighted T_{sk} during summer and post monsoon remained at $35.3 \pm 1.3^{\circ}$ C and $35.0 \pm 1.0^{\circ}$ C, respectively, whereas the value during winter was $31.6 \pm 1.4^{\circ}$ C. One-way ANOVA shows that the T_{sk} of the local areas significantly differed during the three investigating seasons. The trunk, upper arm, hand, thigh, and foot temperatures in summer and post monsoon were relatively more than a winter month.

Different physiological responses, given in **Table 3**, indicates significant differences (p < 0.001) in bodily strains of the stone quarry workers in three seasons of investigation. The average heart rate response of the workers during work in summer and winter seasons were 108 ± 14.6 and 109 ± 21.2 beats/min, respectively, whereas the heart rates were relatively less (99 ± 14.6 beats/min) in the month of post monsoon. The increase in heart

Variable	Statistics	Summer	Post monsoon	Winter		
		(N = 521)	(N = 214)	(N = 199)		
Dry bulb	$\text{Mean}\pm\text{SD}$	40.0 ± 2.4	35.2 ± 2.2	26.6 ± 5.3		
temperature (⁰ C)	5th Percentile	36.1	33.1	20.0		
	95th Percentile	43.2	38.9	34.5		
	Skewness	0.3	0.6	0.2		
	Kurtosis	0.5	-0.9	-1.6		
	F Value	1,265.8 (p < 0.001)				
Outdoor WBGT	$\text{Mean}\pm\text{SD}$	35.5 ± 2.3	32.2 ± 1.8	23.1 ± 2.0		
(⁰ C)	5th Percentile	31.8	28.1	20.0		
	95th Percentile	39.4	35.4	26.8		
	Skewness	0.7	-0.4	0.1		
	Kurtosis	0.6	0.2	-0.7		
	F Value	:	2,382.0 (p < 0.001)		

TABLE 1 | Environmental conditions at workplaces.

rate in winter is due to decrease in environmental warmth and increase in physical load. The environmental load and heart rate responses differed significantly in summer and post-monsoon months of investigation $[F_{(2,931)} = 31.3, p < 0.001]$. About 85% of the workers in summer, 68% in post monsoon, and 79% in winter have working heart rates greater than 90 beats/min. The heart rate response and the prediction of oxygen uptakes at the range of 0.76–1.96 l/min for the workers, indicate that the severity of stone quarry work might be categorized as heavy to extremely heavy in summer and heavy to moderately heavy in post monsoon and winter. The systolic blood pressure in the month of winter was marginally higher (139 ± 15 mmHg) as compared to summer months (133 ± 17.1 mmHg). Also there was no significant difference in diastolic blood pressure during three seasons.

The average level of T_{cr} of the stone quarry workers during work in the months of summer, post monsoon and winter illustrates the dynamic equilibrium of heat transfer supposedly maintained between the body core and periphery, in regulating the buildup of body temperature. As given in **Table 3**, the 95th percentile value of T_{cr} in summer months reached 40.1°C. It was noted that nearly 10% of the workers, the T_{cr} level during their work in summer months crossed the critical limit value of heat tolerance (39°C) and these workers were at unsafe zone of exposure. This remains a challenge to recognize those vulnerable workers who might be at risk of heat disorders. None of the workers during post monsoon and winter crossed the heat tolerance criteria.

The workers had similar demographic and physical characteristics and they were engaged in equivalent nature of work. The physiological demand of work for the workers in the month of May to June was ~14% higher, as compared to the strains during post monsoon and winter. The weighted T_{sk} and T_{cr} of the workers are grouped according to the WBGT range (**Figure 2**). There was a consistent increasing trend of T_{sk} and T_{cr} , however the gradient tend to decrease when the WBGT exceeded 32.9°C. This hallmarks the critical zone where the

TABLE 2 | Local skin temperature profile of workers at workplaces.

Segmental T _{sk}	Statistics	Summer (<i>N</i> = 521)	Post monsoon (N = 214)	Winter (<i>N</i> = 199)
Head (⁰ C)	$\text{Mean} \pm \text{SD}$	35.8 ± 1.2	34.8 ± 1.0	30.7 ± 2.4
	5th Percentile	33.0	33.2	26.8
	95th Percentile	37.4	36.7	34.6
	Skewness	-0.1	1.1	-0.1
	Kurtosis	-0.4	0.1	-0.8
	F Value		264.4 (p < 0.001)	
Trunk (⁰ C)	$Mean \pm SD$	35.1 ± 1.6	34.9 ± 1.2	32.1 ± 1.8
	5th Percentile	32.6	33.0	28.8
	95th Percentile	37.5	36.8	34.7
	Skewness	-0.2	1.2	-0.4
	Kurtosis	0.2	0.1	-0.1
	F Value		282.8 (p < 0.001)	
Upper arm (⁰ C)	$Mean \pm SD$	35.1 ± 2.1	• /	29.3 ± 2.1
	5th Percentile	31.2	33.1	25.3
	95th Percentile	38.2	37.0	32.6
	Skewness	-0.5	1.2	-0.4
	Kurtosis	-0.1	0.1	0.2
	F Value		672.5 (p < 0.001)	
Hand (⁰ C)	Mean \pm SD	35.6 ± 1.3	35.0 ± 1.2	32.3 ± 1.6
(-)	5th Percentile	33.6	33.1	29.4
	95th Percentile	38.0	36.7	34.7
	Skewness	0.2	-0.3	-0.3
	Kurtosis	0.3	0.2	-0.3
	F Value		424.6 (p < 0.001)	
Thigh (⁰ C)	Mean \pm SD	35.6 ± 1.9	35.2 ± 1.3	31.7 ± 1.5
5 (- /	5th Percentile	32.7	33.0	29.1
	95th Percentile	38.9	37.2	34.0
	Skewness	0.3	-0.1	-0.3
	Kurtosis	0.3	-0.4	0.8
	F Value		388.3 (p < 0.001)	
Foot (⁰ C)	Mean \pm SD	35.2 ± 3.0	35.2 ± 1.4	28.9 ± 2.4
	5th Percentile	30.1	32.7	24.6
	95th Percentile	40.0	37.3	32.9
	Skewness	-0.1	-0.3	-0.1
	Kurtosis	0.1	-0.3	-0.1
	F Value	011	448.5 (p < 0.001)	011
Weighted T _{sk}	Mean \pm SD	35.3 ± 1.3	35.0 ± 1.0	31.6 ± 1.4
(⁰ C)	5th Percentile	33.0	33.3	29.2
	95th Percentile	37.4	36.6	33.8
	Skewness	-0.1	0.0	-0.3
	Kurtosis	-0.1	-0.3	-0.3 -0.3
	F Value	-0.4	-0.3 264.4 (p < 0.001)	-0.5

probably the thermoregulation mechanism of the body switched on to maintain the body haemostasis and further control build up of temperature at core so, that the workers body could adapt to the thermal environment. The cascades of event occurs to offload the produced heat is by increasing the cutaneous blood flow that bring the hot blood closer to the external environment

Variable	Statistics	Summer (<i>N</i> = 521)	Post monsoon $(N = 214)$	Winter (<i>N</i> = 199)
Heart rate	$\text{Mean}\pm\text{SD}$	$108 \pm$	99 ± 14.6	$109 \pm$
(beats/min)		14.6		21.2
	5th Percentile	88	80	80
	95th Percentile	135	120	152
	Skewness	0.6	0.9	0.8
	Kurtosis	-0.1	2.4	0.5
	F Value		31.3 (p < 0.001)	
Systolic BP (mmHg)	$\text{Mean}\pm\text{SD}$	133 ± 17.1	128 ± 13.4	139 ± 15.0
	5th Percentile	106	109	115
	95th Percentile	158	153	163
	Skewness	0.3	0.6	0.1
	Kurtosis	1.0	0.4	1.3
	F Value		21.9 (p < 0.001)	
Diastolic	$\text{Mean}\pm\text{SD}$	78±12.7	79 ± 12.8	77 ± 10.7
BP (mmHg)	5th Percentile	60	60	60
(mmg)	95th Percentile	98	101	94
	Skewness	1.7	0.6	-0.6
	Kurtosis	14.7	2.0	1.5
	F Value		1.5 (NS)	
T _{cr} (⁰ C)	$\text{Mean} \pm \text{SD}$	37.3 ± 1.1	36.7 ± 0.4	36.8 ± 0.5
	5th Percentile	36.2	36.1	36.1
	95th Percentile	40.1	37.3	37.8
	Skewness	1.8	0.0	0.7
	Kurtosis	3.1	-0.7	0.3
	F Value		42.6 (p < 0.001)	
Sweat loss	$\text{Mean}\pm\text{SD}$	15.6 ± 1.7	13.7 ± 1.5	6.5 ± 1.8
(gm/min)	5th Percentile	13.0	9.7	3.9
	95th Percentile	18.4	15.8	9.8
	Skewness	0.8	-1.1	0.4
	Kurtosis	1.2	1.4	-0.7
	F Value		2,099 (p < 0.001)	
Predicted tolerance	$\text{Mean}\pm\text{SD}$	65 ± 12.7	83 ± 17.4	199 ± 42.7
time (min)	5th Percentile	46	62	131
- \ /	95th Percentile	88	132	266
	Skewness	-0.1	1.7	0.0
	Kurtosis	0.4	2.8	-0.7
	F Value		2,429.2 (p < 0.001)	

and loose the heat by radiation, convection, and evaporation of sweat into latent heat.

The segmental heat exchanges and the rate of body temperature build up were estimated and arrived at a time duration that corresponded to the limit of tolerance of 39°C and referred to as heat tolerance time (**Table 3**). During the summer season, the tolerance time was significantly less, in comparison to other two seasons. In post-monsoon season, the tolerance time was arrived at $83 \pm 17 \text{ min}$ at WBGT 32.2 $\pm 1.8^{\circ}$ C; i.e., 18 min drop in tolerance time for 3.3° C increase

in WBGT. For the winter season, the heat tolerance time was estimated as 199 \pm 43 min at WBGT 23.1 \pm 2.0°C. About 134 min drop in tolerance time for ~12°C increase in WBGT in summer season.

The questionnaire surveyed the checkpoint that looked into the signs and symptoms of heat-related illnesses, as given in **Table 4** (Nag et al., 2013; Hanna and Tait, 2015). Corresponding to observations of physiological and subjective responses to heat stress, the workers were vulnerable to heat illnesses. Over 21.3% of the stone quarry workers complained of decreased urine output situation during the summer exposure, in comparison to only 7% workers in post monsoon and 17.6% in winter. Nearly 93.5% of the workers complained of excessive sweating and 88.7% feeling of excessive thirst/dry mouth and ~58.7% workers reported elevated T_{cr} during the summer months. About 3/4th of the workers complained of decreased working capacity.

Perceived effort/exertion of an individual scored using Borg's scale corresponded closely to the severity of the tasks performed. The perceived effort levels remained in the range of 14-17 and for this level of subjective response, the heart rate variations might correspond to 140-170 beats/min. However, the 95th percentile values of heart rates for the workers in the months of summer, post monsoon and winter were 135, 120, and 152 beats/min, respectively. The subjective response to overall physical fatigue score remained at a high level, i.e., close to 9-10 in 13 point scale, however, the relative fatigue to different levels of environmental warmth could not be reflected. The self-reporting of perceived effort, physical fatigue, and any other heat-related symptoms by the illiterate workers have limitations and therefore, appropriate indoctrination of the workers and consistent recording by the field investigators was essential in establishing relationships between the symptoms and heat exposures.

Our study shows a need of break or rest rooms of the workers that may effective in reducing the thermal load of participants. Further, studies on stone quarry work may be advantageous in estimating the exact nature of thermal load experienced by workers and its discernible effects. Nevertheless, it will help in understanding India's burden of heat stress illness, both occupational and otherwise.

DISCUSSION

The stone quarry work as compared to other outdoor thermal environment occupation like steel plant, steel plant, power plant, and forge plant where it is difficult to control the environmental adversaries which becomes confounding variables in the study of such occupations. According to Indian Meteorology Department (IMD), a category of heat wave includes places where the normal maximum temperature is more than 40°C. Researchers had found that for the last 25 years the average global temperature rose by 0.6° C (De et al., 2005; IPCC, 2007). The conditions of the heat wave prevailed in the regions where the study was undertaken during the summer (May to June), as the 95th percentile value of dry bulb temperature was 43.2° C. The



environmental load in the month of winter was substantially less in comparison to conditions during the season summer and post monsoon.

The cardiovascular and thermoregulatory responses of the stone quarry workers differed significantly in the month of summer, post monsoon, and winter. The responses were the resultant of combined effect of environmental warmth and work strenuousness that ranged from heavy to extremely heavy in the month of summer, post monsoon and heavy to moderately heavy in the month of winter. The study observed a small increase in systolic blood pressure during the winter months. The trend of the results corroborates to the findings of Kristal-Boneh et al. (1995) that the average Systolic BP at work was higher in winter than in summer. The activation of the sympathetic nervous system and secretion of catecholamine might be increased in cooler environment, resulting in increase in blood pressure through an increased heart rate and peripheral vascular resistance (Alperovitch et al., 2009).

The relative effects of environmental stress on the physiological responses that would be expected beyond the level attributed to physical work, however, need to be ascertained. Data indicated that the environmental effects on local segmental T_{sk} and weighted average T_{sk} of workers were greater than the

effects that might be attributed to work severity (Nag et al., 2013). The profile of segmental T_{sk} indicated deviation from the thermo-neutral reference, provoking distinctive peripheral response for feedback and regulation in building up of body temperature. For the range of environmental warmth from 25 to 43°C WBGT (ISO Standard 7243, 1989), the workers had an increasing trend of T_{sk} and T_{cr} , however the gradient tended to narrow down when the WBGT exceeded 32.9°C and the gradient was found to be < 3°C (Nag et al., 1997, 2013). The stone quarry works are performed directly under sun and the physiological demand of work in the month of summer was ~14% higher, as compared to the demands in the months of post monsoon and winter.

However, the biophysical analysis of heat exchanges between the body core and skin surface yield the rate of body core temperature build up and accordingly, the tolerance time of heat exposure was arrived at, corresponding to the of T_{cr} 39°C (Hanna and Tait, 2015). Above 39°C of T_{cr} , serious heat stroke and neurological effects may occur to a worker (Parsons, 2003).

As observed, there was considerable difference in the tolerance time of stone quarry work in three different seasons, due to the differences in the environmental variables and workload. The tolerance time level in summer months (65 \pm 13 min at WBGT 35 \pm 2.3°C) was less than other two seasons (post monsoon and

	Summer (N = 521)	Post monsoon $(N = 214)$	Winter (N = 199)	
	% of workers expressed heat strain			
Heavy sweating	93.5	91.1	69.8	
Elevated heart rate	76.0	59.8	61.3	
Weakness or fatigue	75.2	78.5	62.3	
Dizziness/nausea	40.5	31.8	41.2	
Headache	51.1	43.5	44.2	
Confused and irritated	44.5	20.1	19.1	
Skin tanning	55.1	9.8	23.6	
Excessive thirst/dry mouth	88.7	82.7	55.3	
Decreased urine output	21.3	7.0	17.6	
Loss of appetite	47.0	26.6	36.7	
Blurred vision	44.5	37.4	25.1	
Hot or dry skin (no sweating)	21.9	7.0	18.1	
Red face	51.2	36.9	20.1	
Chill feeling/shivers	45.5	31.8	23.6	
Mental disorientation	43.8	15.4	27.6	
Elevated body temperature	58.7	17.8	52.3	
Seizure	13.4	0.0	6.5	
Slurred speech	7.7	0.5	4.0	
Abdominal spasms	35.9	32.7	31.2	
Muscle pain/cramp (arms/legs)	48.2	57.9	64.3	
Fainting/feel collapse	13.6	4.7	28.6	
Pink or red bumps	35.3	25.2	10.1	
Itching skin	39.2	24.3	14.1	
Irritation or prickly sensation	29.6	31.8	13.1	
Loss of work capacity	75.6	62.1	57.3	

winter). From the cross-sectional data on stone quarry workers, it was estimated that there was \sim 14% loss of tolerance time per degree increase of WBGT, from 33 to 35°C WBGT. The loss of tolerance time might also indicate loss of productivity due to heat exposure, which Kjellstrom (2016) referred to as High Occupational Temperature Health and Productivity Suppression (Hothaps) effect, for loss of working ability or working capacity. The relative workload was higher during winter season. It is likely that the workers might be adopting self-adjustment strategy in the pace of work distributing the work and workload as per the varying environmental exposures. The make-shift shelters where the workers take rest during the hottest hours. It was observed that the environmental effects on workers appeared to be greater than the effects of work severity, therefore consistent field investigators was essential in establishing relationships between the symptoms and heat exposures.

In repeated occupational exposures high heat load and strenuous physical activity, human's defense mechanism undergoes progressive changes for internal thermal stability (acclimatization), depending upon on physiological adaptive capacity (Morioka et al., 2006). Data amply suggest that the workers during the summer months were at unsafe zone of exposure and 14% of the workers were vulnerable to heat illnesses. Also, the workers lack awareness and measures to mitigate risks. This kind of data from a larger sample size is greatly important in the assessment of the health, safety and productivity impacts of climatic changes with seasonal variation, and therefore, might be useful to develop prevention programmes of the population at risk to heat waves.

STRENGTHS AND LIMITATIONS

The location of this cross-sectional study of stone quarry workers was the same in summer, post monsson and winter months but, the specific workers and worker tasks differed between the survey times. However, regardless of the possible difference in three seasons the survey participants and their ativities, perceived heatrelated symptoms and environmental measurments of heat stress has supported our overall finding that heat stress is an important risk factor for worker health. Also of note is that this study may be conservative in its findings as responses related to heat disorders among stone quarry workers may be higher than those figures observed in the study due to a healthy worker bias (i.e., those most affected by the heat were absent or had stopped doing this type of work). Another possible explanation may be attributed to the fear of being reprimanded by the management for discussing issues that may portray them negatively.

A key strength of this study is that we surmised that the high exposure coupled with strenuous physical load are the major contributing factors. Further, there is lack of Indian heat exposure guidelines for determining ceiling limits of environmental exposure for tropical heat exposure of the population. Our study supports the establishment of separate tropical or India specific heat exposure guidelines and interventions that could simultaneously be worker protective but realistic in this climate.

CONCLUSION

The study bears considerable practical importance to assess the magnitude of thermal stress among stone quarry workers in the working environment and the worker's physiological reaction to it, and therefore to ensure optimal conditions for health and productivity. The study has a limitation of focusing only on three seasons, however, the basic premise was that the cardiovascular and thermoregulatory parameters are critical to manifest ones thermal environmental perceptive responses in a occupational situation. The habitual occupational involvement makes the workers naturally acclimatized to hot environment, however, even the habitual workers during peak summer months are at potential risk of developing heat-related illness. The comprehensive analysis of the physiological and thermoregulatory responses of workers to heat stress and strain would eventually ascertain the relative vulnerability of the stone quarry workers for their exposure to extreme hot environment.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors if required, without undue reservation. But, in that case the identity or the personal information of the participants will not be shared.

ETHICS STATEMENT

The studies involving human participants were reviewed and approved by Indian Council of Medical Research. The

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patients/participants provided their written informed consent to participate in this study.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct and intellectual contribution to the work, and approved it for publication.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Elemental Characteristics and Source-Apportionment of PM_{2.5} During the Post-monsoon Season in Delhi, India

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Bangar V, Mishra AK, Jangid M and Rajput P (2021) Elemental Characteristics and Source-Apportionment of PM_{2.5} During the Post-monsoon Season in Delhi, India. Front. Sustain. Cities 3:648551. doi: 10.3389/frsc.2021.648551 In this study, we have coupled measurements, modeling, and remote sensing techniques to better delineate the source characteristics and variability of air pollutants in Delhi primarily during the post-monsoon season in 2019. We show a comparison of ambient $PM_{2.5}$ (particulate matter having aerodynamic diameter $\leq 2.5 \,\mu$ m) levels and associated elements during the post-monsoon with those during a relatively clean season of monsoon (experiencing frequent wet precipitation). Air-mass back trajectories from Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model have been used to infer the possible source pathways of PM_{2.5} impacting at the receptor site in Delhi. The average concentrations of PM_{2.5} during monsoon (June–July) and post-monsoon (October-November) were 42.2 \pm 15.5 μ g m⁻³ (range: 22-73 μ g m⁻³) and 121.4 \pm 53.6 μ g m⁻³ (range: 46–298 μ g m⁻³), respectively. The PM_{2.5} samples were analyzed for heavy and trace elements (Si, S, Na, Mg, Al, Cl, Ca, K, Ti, V, Cr, Mn, Fe, Ni, Cu, Br, Rb, Zr, and Pb) using an Energy Dispersive X-ray Fluorescence (ED-XRF) technique and their concentrations have been used to carry out the source-apportionment utilizing principal component analysis (PCA) tool. The PCA analysis has identified three major sources of fine aerosols including contributions from the sources viz. vehicular emission, biomass burning, coal combustion, secondary aerosols formation, soil dust, solid-waste burning and industrial emission. The source involving biomass burning contributed largely to the PM_{2.5} in post-monsoon season through long-range transport of large-scale agricultureresidue burning emissions (occurring in the states of Punjab, Haryana, and western part of Uttar Pradesh). The industrial emissions include primarily, medium- and small-scale metal processing industries (e.g. steel sheet rolling) in Delhi-National Capital Region. Traces of emission from coal based thermal power plants and waste incineration have also been observed in this study.

Keywords: atmospheric aerosols, trace metals, urban air-shed, source-apportionment, Delhi

INTRODUCTION

Clean and healthy air is essential to all life on earth and is crucial for the well-being of human beings and the optimum performance of its supporting ecosystems. However, industrialization and urbanization have severe detrimental effects on the natural environment, be it air, water, or the soil (Kushwaha et al., 2012). The atrocious levels of particulate matter (PM) pollution in the atmosphere has created a disconcerting situation across the world's scientific and political communities, especially in developing countries due to their climatic and human health impacts (Khodeir et al., 2012; World Health Organization, 2016). PM pollution alters the composition and chemistry of the lower atmosphere, degrades air quality, reduces visibility and impacts the global climate (Khain and Pinsky, 2018). Numerous harmful impacts of PM exposure such as pulmonary and cardiovascular diseases, allergies and premature deaths have been evinced in several epidemiological studies (Badyda et al., 2016; Ghude et al., 2016). A recent research has reported that more than 75% people in India are imperiled with $PM_{2.5}$ levels >40 µg m⁻³, a limit set by the National Ambient Air Quality Standards (Balakrishnan et al., 2019). The study has also mentioned that exposure to ambient PM2.5 resulted in ~9.8 lakhs premature deaths in a year and is of the major concern to human health point of view. Another research has estimated that PM2.5 exposures could lead to average loss of life expectancy (LLE) of 3.4 years for the country with the highest value of LLE of 6.3 years for Delhi (Ghude et al., 2016).

Recent studies have established that the impact of $PM_{2.5}$ on human health cannot be only connected directly to the total mass concentration but also to the toxicity of particulate matter constituents (Yadav and Phuleria, 2020), specifically trace metals (Lippmann and Chen, 2009; Stanek et al., 2011). It is also imperative to analyze the seasonal variation of $PM_{2.5}$ in the ambient air due to its high association with meteorological parameters such as relative humidity, wind speed, temperature, and precipitation (Das et al., 2020). Thus, a good understanding about chemical composition and emission sources of PM along with its seasonal variability is essential for attributing its major impacts and developing effective and efficient mitigation policy strategies (Bangar et al., 2020).

PM2.5 has myriads of natural and anthropogenic sources and is known to originate from the combustion of fossil fuels, biomass burning, soil dust, coagulation of ultrafine particles, reactions in water droplets, and condensation of gaseous organic and inorganic molecules, among others (Sioutas et al., 2005; Edgerton et al., 2009). Studies on elemental composition associated with fine PM in India have revealed that the urban population is particularly exposed to elevated levels of elements including Na, K, Mg, Al, Ca, S, Si, Cl, Cr, Ti, As, Br, Pb, Fe, Zn, and Mn due to vehicular emissions, biomass burning, industrial emissions, and soil dust uplift (Kulshrestha et al., 2009; Murari et al., 2015; Sharma and Mandal, 2017). The extent of health impact and toxicity of PM varies as a function of its composition and source, thus making it essential to conduct source apportionment analysis to comprehend the sources contributing to the PM budget over a receptor site (Jain et al., 2020).

Receptor models use a multivariate statistical approach to identify and quantify the sources of air pollutants, assuming the mass conservation between the emission sources and receptor site (Hopke et al., 2006). In India, source apportionment studies for PM have been performed using different receptor models viz. chemical mass balance (CMB) (Sharma and Patil, 1994; Srivastava et al., 2005; Srivastava and Jain, 2008; Gummeneni et al., 2011), positive matrix factorization (PMF) (Jain et al., 2020; Tobler et al., 2020), and principal component analysis (PCA) (Karar and Gupta, 2007; Suman and Pal, 2010; Hazarika et al., 2015). Valuing the importance and possibility of identifying PM sources, the present study has been conducted using PCA. PCA can proficiently perform the source apportionment analysis without any prerequisite for source profile of constituents associated with PM (Karagulian and Belis, 2012).

The capital city of India (Delhi) is one of the most populous and drastically polluted cities on earth (Bhat, 2020). The rapid rise in demands for housing and infrastructure, production and manufacturing industries, motorized vehicles, and lack of adequate air pollution control programmes have exaggerated the health risks due to PM exposures in the city (Das et al., 2020). Sharma et al. (2016a), using a receptor model technique, identified that transport sector, biomass burning, and industry are the major contributors of PM2.5 in Delhi during the winter season. For summer season, Sharma and Mandal (2017) identified long-range transport of soil dust from regional and transboundary areas as the key contributor of PM_{2.5}. Several other studies have also presented quite similar results; however, comprehensive studies comparing seasonal variation in PM concentrations, and elemental composition as well as confirming the sources of PM pollution (using modeling and remote sensing techniques) in India are scarce. The present work is an effort to distinguish the major sources of PM pollution in Delhi by incorporating measurements, modeling and remote sensing techniques with an aim to assist policymakers in developing improved and enhanced pollution control strategies to curb PM pollution. The current study emphasizes on analyzing the seasonal variations (monsoon and post-monsoon season) in the elemental composition of PM_{2.5} and its source-apportionment during post-monsoon season in the year 2019. Samples of PM2.5 were collected at Jawaharlal Nehru University, New Delhi, India and were assessed for elemental analysis using EDXRF (Energy Dispersive X-Ray Fluorescence). It is an extensively utilized method for the quantification of trace elements in airborne PM due to its nondestructive technique which provides elemental composition results quickly without any chemical pre-treatment (Nascimento Filho, 1999; Zucchi et al., 2000; Öztürk et al., 2011). The measured elemental composition was then used as an input to run the PCA model to quantify the sources contributing to PM_{2.5} primarily for the post-monsoon season. Due to limited number of samples (n = 20) the PCA analysis was not performed for the monsoon season dataset. Moreover, the air-mass backtrajectories were also utilized in the study to understand the impact of distant sources regions on the PM2.5 loading at the receptor site.

METHODOLOGY

Sampling Site

Samples of PM2.5 were collected at Jawaharlal Nehru University (JNU), New Delhi (28.539°N, 77.167°E), India on the rooftop (16 m height) of the School of Environmental Sciences (SES) in the year 2019. The map of the sampling site is shown in Figure 1 (source: Google maps and ArcGIS Pro version 1.2.0) and the locations of all the sites (sampling site and CPCB sites) have been marked. JNU is located in the south-west region of Delhi, in an environmentally sensitive area and extends over a large space of natural vegetation in an area of ~800 acres. The sampling site is distant from any principal industrial activities; however, it is surrounded by major roads with high traffic density. Inside the campus the traffic density is relatively very low. The major landform features around Delhi are the Himalayas lying in northnortheast of Delhi approximately at a distance of 160 km, the Thar Desert of Rajasthan lying in the west and the alluvial plains in the south and east of Delhi. Delhi, with a population of more than 16 million people and over 10 million registered vehicles (Rai et al., 2020) remains choked with heavy air pollution, especially in the winter season due to temperature inversion and shallow boundary layer height which entraps a massive amount of pollutants near the ground level (Hazarika et al., 2015).

PM Sample Collection and Analysis

PM_{2.5} sampling during the monsoon (21 June−13 July, 2019; n = 20) and the post-monsoon (11 October−20 November, 2019; n = 40) seasons was done using a Fine Particulate Air Sampler (APM 550, Envirotech, India). The mean flow rate of the sampler was maintained at 1 m³ h⁻¹ (accuracy ±2%). The sampler was operated for 24 h (8:00 a.m.−8:00 a.m.), and the volume of the air filtered during each sample collection was used to deduce the mass concentrations of PM in the ambient atmosphere. Standard protocol for the sample collection and storage until chemical analysis has been followed as prescribed by the Central Pollution Control Board (CPCB), India.

The samples were collected on the hydrophobic PTFE filter (Fluoropore, FHUP04700) of 47 mm diameter. The moisture of the filters (before and after sampling) was removed by desiccating them in a silica based desiccator. The initial and final weight of the filter substrate was determined on a microbalance and the $PM_{2.5}$ concentration was calculated using the standard gravimetric method. The meteorological data of temperature, humidity and wind speed was taken for both the seasons from Safdarjung airport (IMD: Indian Meteorological Department) site.

For the quantification of metals in PM_{2.5}, an Energy Dispersive X-ray fluorescence spectrometry (ED-XRF) was used for its non-destructive mechanism and quick analysis (Khodeir et al., 2012; Moriyama et al., 2014; Shaltout et al., 2017). In the present study, a total of 19 heavy and trace elements (Si, S, Na, Mg, Al, Cl, Ca, K, Ti, V, Cr, Mn, Fe, Ni, Cu, Br, Rb, Zr, and Pb) were analyzed using ED-XRF (PANalytical Epsilon 5 analyzer). The quality control and analysis (QC and QA) have been carried out during the elemental analysis on ED-XRF. The apparatus was set up with standard reference material (NIST SRM 2783). The

instrument was pre-calibrated using the XRF standard BRPC3 and it performs a semi quantitative analysis. Semi-quantitative analysis allows the users to compare spectral data from samples in order to ascertain relative elemental concentration between samples. Accordingly, the uncertainty in the concentration of elements on ED-XRF was found to be within 10%. The lower limit of detection (LLD) of each element for ED-XRF is provided in **Supplementary Table 1**. The description of elemental analysis using ED-XRF and detailed methodology for calculating the concentration of individual elements are provided in the previous literatures (Maciejczyk et al., 2005; Hazarika et al., 2015). In the present study, the concentration of each element in ng/m³ was calculated using the following equation (Hazarika et al., 2015):

Conc. of element x in ng/m³
=
$$\frac{\text{EDXRF value of x element } \times \text{(Difference in filter weight)}}{\text{Volume of the air sampled}}$$

where EDXRF value denotes the concentration of element x in ppm, difference in filter weight (pre- and post-sampling) is in milligrams and the volume of air sampled is in m^3 .

Air-Mass Backward Trajectory Analysis

The fine PM has relatively a high residence time, and it can undergo long-range transport in the ambient atmosphere (Maenhaut et al., 2016). Therefore, to understand the origin of PM_{2.5} and its long-range transport pattern to the sampling site, the air-mass backward trajectory analysis was done using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Daily GDAS (1 degree, global) meteorological files were accessed to compute the 72-h back trajectories at 500 m and 1,000 m above the ground level (AGL) for each day and night of the sampling campaign. These heights were chosen to represent winds in the boundary layer and to eliminate local land cover and topographic effects. AIRS (Atmospheric Infrared Sounder) dust score data from NASA website (URL: https://earthdata.nasa.gov/labs/worldview/) and MODIS Moderate Resolution Imaging Spectroradiometer fire count data (URL: https://earthdata.nasa.gov/active-fire-data) was overlaid on a true color image to locate the geographical regions responsible for enhancing the aerosol concentrations at the sampling site. AIRS dust score layer shows the level of dust aerosols in the Earth's atmosphere and the areas affected by it. Whereas, the MODIS thermal anomalies data is a fire product in which active fires and other thermal anomalies are identified using thermal anomalies algorithm (Giglio et al., 2003).

Source Apportionment Using PCA

The source apportionment analysis of PM_{2.5} was carried out only for the post-monsoon season using the Principal Component Analysis (PCA). PCA is a statistical tool which explains the variance of a large amount of data having inter-correlated variables and transforms it into a smaller dataset of independent variables called principal components (PC) (Thurston and Spengler, 1985; Sharma et al., 2016b). In PCA, the factor loadings or PC identify the sources associated with pollution based on the correlation of individual pollutant species with each component.



The pollutant species highly correlated with individual PCs indicates the association of that PC with the source emission composition (Johnson et al., 2015). In the present analysis, PCA has been performed with a statistical software SPSS (Version 25) following the Varimax Rotation method. This method maximizes the variance of the squared elements in the column of a factor matrix. In PCA, the first PC explains the most significant fraction of the original variables, while the second PC estimates a reduced fraction of the original variable in comparison with the first PC and so on (Sousa et al., 2007). PCA begins by normalizing the set of variables as Z_{ij} using equation (1), so that the variance of this set of variables is unity.

$$Z_{ij} = \frac{C_{ij} - \overline{C_j}}{\delta_j} \tag{1}$$

Where C_{ij} is the concentration of jth species in the ith sample; $\overline{C_j}$ and δ_j are the average concentration and standard deviation of that species j. The fundamental operation of PCA can be

expressed by equation (2) indicating that it splits the data matrix into two matrices G_{ik} (factor loading) and H_{kj} (factor score), as shown below:

$$Z_{ij} = \sum_{k=1}^{p} G_{ik} H_{kj} + E_{ij}$$
(2)

Here, i, j, and k represent the index for sample, species and factors, respectively. E_{ij} represents the residual matrix. The two vectors G and E are unknown in the factor analysis (FA) and are obtained by assuming various covariance relationships between the vectors H and E and finally Varimax rotation of matrix is applied to minimize the datasets of elements having high loading factor (Kumar et al., 2001). As the factor load of the variable increases, the identification of the possible source of components also increases (Henry, 2003). The detailed methodology for air pollutant's source identification and apportionment using PCA is provided in Chavent et al. (2009). In the present study PCA was carried out utilizing elemental concentration data of 40

samples for the post-monsoon season. Numerous other studies have employed PCA for source apportionment using limited data points viz; Hazarika et al. (2015) utilized data from 12 samples for each season (summer, winter and monsoon) to perform PCA analysis, Liu et al. (2018) utilized data from 15 day samples, similarly Zhang et al. (2019) and Kanellopoulos et al. (2020) also utilized limited data set to carry out source apportionment analysis using PCA.

RESULTS AND DISCUSSION

Seasonal Variability of PM_{2.5} and Air-Mass Back Trajectory Analysis

In total, 60 samples were collected to ascertain the seasonal variation in $PM_{2.5}$ concentrations during monsoon (n =20 samples) and post-monsoon (n = 40 samples) seasons. Average mass concentrations were found to be 42.2 \pm 15.5 μ g m⁻³ (range: 21.8–72.5 μ g m⁻³) and 121.4 \pm 53.6 μ g m⁻³ (range: 45.9-298.1 µg m⁻³) in monsoon and post-monsoon seasons, respectively. The PM2.5 data monitored by the Central Pollution Control Board (CPCB) at four nearby sites (Aya Nagar, IGI Airport, PUSA and Sri Aurobindo Marg) was also taken into account to assess the spatial heterogeneity in PM2.5 mass concentrations. Figure 2 shows the temporal variability of PM_{2.5} concentrations at the sampling site along with the mean concentration for the aforementioned nearby CPCB sites. The average PM_{2.5} concentration at the sampling site i.e., JNU is lower than the other adjoining areas particularly in the post-monsoon season. The temporal variation of PM2.5 mass concentrations for all sites during both the seasons are provided in Supplementary Figure 1. The average PM_{2.5} mass concentrations at JNU and that integrated for the four CPCB sites were 42.2 μ g m⁻³ and 44.3 μ g m⁻³ in the monsoon season and 121.4 μ g m⁻³ and 183.3 μ g m⁻³ in the post-monsoon season. This can be attributed to the vast tract of natural vegetation and the low traffic density in the University campus site (JNU).

Figure 3 shows the daily variation of meteorological parameters (temperature, humidity and wind speed) during both the sampling seasons. It is apparent from the figure that the monsoon season was warmer than the post-monsoon season. The wind speed in the monsoon season was also higher than the post-monsoon season. However, the relative humidity appeared to be higher in the post-monsoon season as compared to the June-July period of the monsoon season. It is evident from Figure 2 that the mean concentration of $PM_{2.5}$ was substantially higher in the post-monsoon season than the monsoon season. The previous studies conducted in Delhi assessing seasonal variations in PM_{2.5} also showed an increased concentration in post-monsoon and winter season with respect to the monsoon season (Mandal et al., 2014; Gopalaswami, 2016; Panda et al., 2016; Sharma and Mandal, 2017; Jain et al., 2020). The most plausible explanation for lower PM2.5 mass concentrations during the monsoon season relates to the high ventilation and dispersion of pollutants as well as occurrence of frequent precipitation leading to washout of the air pollutants during the monsoon (Jain et al., 2020). In the post-monsoon season, activities such as stubble (agriculture residues) burning in the fields of Haryana, Punjab, and western part of Uttar Pradesh, firecrackers burst during Diwali festival along with prevailing meteorological conditions such as minimal wind speed, shallow boundary layer height, and geographical settings (Himalayan range in the north and Deccan Plateau in the south) results into entrapment of a large amount of PM_{2.5} near the ground level over the study region (Perrino et al., 2011; Rai et al., 2020). Kulshrestha and Kumar (2014) in their review report highlighted the need and significance of trajectory analysis for identifying the sources of PM pollution in South Asia.

The results from HYSPLIT back trajectory analysis indicating for the long-range transport of air masses at the sampling site are illustrated in Figures 4A-E. Air-mass back trajectories for a particular season do not look very different at two different altitudes, i.e., 500 and 1,000 m. During the monsoon season, the 72-h back trajectory indicated that the air masses traveled longer distances due to higher wind speed (Figures 4A,B). Most of the air masses in this period advanced to the receptor site from Indo-Gangetic Plain (IGP), Rajasthan, Gujarat, Pakistan, and the northern Arabian Sea, along with traces of air masses from Middle-East and the northern Bay of Bengal. The AIRS dust score overlaid on a true color image from MODIS shown for 24 June 2019 (Figure 4C) indicated for a strong dust layers in the atmosphere over Pakistan, Middle East and parts of the Arabian Sea which indicates that maybe the dust particles from these places would have transported over the receptor site. However, clean air mass from the Arabian Sea and Bay of Bengal would have diluted the overall dust load impact from these places at the receptor site. Sharma and Mandal (2017) made similar observations with the air-mass trajectory analysis during the monsoon season of 2013 over Delhi. Sharma et al. (2010) in their study of SO₂ variation over Delhi also highlighted the long-range transport of air-masses from sources pointing toward western and southwestern regions in the monsoon season.

In the post-monsoon season (Figures 4D,E), the backtrajectory showed that the air masses have covered shorter distance, attributable to the lower wind speed, before arriving at the receptor site. The air masses in this period were approaching the receptor site mainly from states of Haryana, Punjab, IGP region, Rajasthan, and Pakistan. The MODIS fire count data plot (Figure 4F) for 04 November 2019 exhibits an intensive stubble burning in Punjab, Harvana and western part Uttar Pradesh indicating that high concentration of PM (and gaseous pollutants) from biomass burning emission could severely impact the air quality at the receptor site. Moreover, emissions from numerous industries operating in Delhi would also contribute to PM loading at the receptor site. Therefore, higher concentration of PM_{2.5} in the post-monsoon season can be attributed to the combined effect of lower wind speed, shallower boundary layer height, and the high incidence of stubble burning which increases the aerosol load over Delhi (Kanawade et al., 2020). For post-monsoon season similar results mentioning stable meteorological conditions leading to accumulation of local and transboundary pollutants have been mentioned in the previous studies conducted in Delhi (Tiwari et al., 2013; Panda et al., 2016; Cusworth et al., 2018; Kulkarni et al., 2020; Nair et al., 2020).



In sharp contrast, the precipitation and high wind speed lead to lowering the concentration of atmospheric particles during the monsoon season (Sharma et al., 2016a). It is worth mentioning that the aerosols sampling was performed for 60 days, spread over both the seasons, and we found that the PM concentrations exceeded the NAAQS (National Ambient Air Quality Standard) standard value for more than 70% of the days. This is coherent with the other research work carried out over the Delhi (Pachauri et al., 2013; Tiwari et al., 2013; Sahu and Kota, 2016; Das et al., 2020; Jain et al., 2020).

Elemental Composition of PM_{2.5}

Figure 5 shows the percentage elemental composition of fine particulates for both seasons [(a) monsoon and (b) post-monsoon]. The daily variations in the percentage elemental composition of PM_{2.5} for both the seasons are shown in **Supplementary Figure 2**. The temporal variations in concentration of the analyzed elements associated with PM_{2.5} for both the seasons are depicted in **Supplementary Figures 3A–C**. The result of the current study showed that the average concentrations of elements were found in the decreasing order of Si>Al>Na>Mg>S>Ca>Cl>K>Fe for the monsoon season and S>Al>Na>Si>Cl>K>Mg>Ca>Fe for the post-monsoon season. The presence of Si, Al, Na, Mg, S, Ca, Fe and K as major elements associated with PM_{2.5} is in agreement with other

studies conducted in Delhi by Jain et al. (2017) and Sharma and Mandal (2017). Hazarika et al. (2015) also observed Na, Ca, Si and K as abundant elements in PM2.5 followed by Ni, Cu and Pb. However, in the present study, the mean elemental concentration of individual elements was comparatively less than the previous studies conducted in Delhi. The relative contribution of elements associated with crustal or natural origin such as Si, Al, Na and Mg (Pipal et al., 2014; Ali et al., 2019; Rai et al., 2020) accounted for 87% of the elemental composition of PM_{2.5} in the monsoon season, whereas in the post-monsoon season these elements accounted only for 57% (Figure 5). The element which contributed the most to the fine particulate was Silicon (Si) 39% in the monsoon season and Sulfur (S) 30% in the post-monsoon season. The high concentration of Si could be attributable to uplifted mineral dust contribution, whereas elevated S content may have contribution from coal combustion and agricultural-residue (biomass) burning (Sternbeck et al., 2002; Perrino et al., 2011). The sources of soil/road dust in the atmosphere include transboundary transport from deserts or entrainment from paved or unpaved roads, construction activities, and agricultural practices (Kulshrestha et al., 2009; Tiwari et al., 2013).

Moreover, the contribution of Potassium (K), Chlorine (Cl), and Lead (Pb) have been found to increase in the post-monsoon season due to different reasons. Higher concentration of K can



FIGURE 3 | Daily variation of meteorological parameters; (A,B) air temperature (in °C), (C,D) humidity (in %), and (E,F) wind speed (in mph) during the monsoon (21 June –13 July 2019) and post-monsoon (11 Oct–20 Nov 2019) seasons, respectively.



FIGURE 4 | HYSPLIT derived air-mass back trajectories during (A,B) monsoon (21 Jun–13 Jul 2019) season and, (D,E) post-monsoon (11 Oct–20 Nov 2019) season at 1,000 m and 500 m. Day time and night time wind trajectories are shown by red and blue colors, respectively. (C) MODIS aqua true color imagery overlaid with AIRS dust scores for 24 Jun, 2019 (major wind direction at receptor site are shown by arrows). (F) MODIS aqua true color imagery overlaid with MODIS fire counts for 4 Nov, 2019 (major wind direction at receptor site are shown by arrows).

be attributed to local biomass burning (in addition to some fraction from upper continental crust) for space heating and agricultural residue burning (Liu et al., 2018; Das et al., 2020);

Cl can have contributions from lubricants, diesel fuels, coal combustion, biomass burning, and plastic and paper burning (Singhai et al., 2017; Chang et al., 2018); Pb to ore and metal



processing, lead-acid battery production/recycling as well as waste incineration (Bukowiecki et al., 2009; Kothai et al., 2011). Summing up, the post-monsoon season witnessed a substantial increase in PM2.5 concentrations predominantly due to the anthropogenic emissions (as suggested by elevated levels of S, K, Cl, Pb) with a relatively low contribution of mineral dust as compared to the scenario in monsoon season. Similar remarks have been noticed before in the previous research works (Khodeir et al., 2012; Das et al., 2020; Jain et al., 2020; Rai et al., 2020). Within the trace elements, three carcinogenic heavy metals were identified, i.e., Ni, Cr and Pb, that may pose substantial risk to humans (Liu et al., 2015). The estimated concentration of these elements is depicted in Supplementary Figure 3B. However, their concentration in this study was found well within the limits prescribed by the World Health Organization (WHO). The comprehensive details on source apportionment of PM2.5 based on associated metals profile are discussed in the following section.

Source Apportionment

To distinguish the possible sources of fine fraction particulate matter, the principal component analysis (PCA) was carried out. Based on the eigenvalues >1, PCA segregates the data into several clusters known as principal components (PC), which also indicate the contribution of each dependent variable (which in this study is the element concentration) in terms of factor loadings. The PCA was performed using the data set consisting of 19 elemental species in 40 PM_{2.5} samples (during the postmonsoon season) collected at JNU site in Delhi. **Table 1** shows the output of PCA analysis using SPSS (IBM, SPSS, version 25) software for the post-monsoon season. The correlation matrices for all 19 elements during both the seasons are shown in **Supplementary Tables 2, 3**. **Figure 6** shows the possible sources of PM_{2.5} pollutions in the post-monsoon season as identified

 $\begin{array}{c} \textbf{TABLE 1} & \textbf{Summary of principal component analysis (PCA) of elements} \\ associated with PM_{2.5} \mbox{ over Delhi during the post-monsoon season.} \end{array}$

Species	Post-monsoon season		
	PC1	PC2	PC3
Na	0.625	0.532	0.490
Mg	0.835	0.460	0.165
Al	0.924	0.322	0.104
Cu	0.414	0.342	0.822
Ni	-0.009	0.795	-0.067
Zr	0.120	-0.155	0.692
Pb	0.140	0.738	0.049
Rb	0.302	0.320	0.815
Br	0.132	0.325	0.840
Fe	0.442	0.852	0.158
Mn	0.350	0.850	-0.101
Cr	-0.273	0.298	-0.473
V	0.953	0.046	0.050
Ti	0.961	0.172	0.081
Si	0.829	0.468	0.150
S	0.536	0.725	0.276
Са	0.694	0.589	0.247
CI	-0.191	-0.106	0.761
К	0.888	0.227	0.297
Variance (%)	33.66	27.42	20.55
Cumulative Variance (%)	33.66	61.08	81.63

The bold value shows the factor loadings greater than 0.5 indicating the inclusion of those elemental species into each Principal Component (i.e. PC1, PC 2, PC3).

using PCA. The total variance explained for the sources of $PM_{2.5}$ during post-monsoon season (81.7%) using PCA are provided in **Supplementary Table 4**.


During the post-monsoon season, the PCA analysis revealed three factors, accounting for 81.7% of the total variance. The first PC explained 33.7% of the overall variance with an eigenvalue of 6.39 with high loadings of Na, Mg, Al, K, Ca, Ti, V and Si. Perrino et al. (2011) and Sharma et al. (2016a) reported that crustal dust is the major source of Si, Na, Mg, Ca, Ti, and Al in particulate matter. Therefore, this factor can be wellassociated with crustal suspension. The % contribution of these elements (Figure 5) to fine particulate matter was relatively low in the post-monsoon season with respect to the monsoon season possibly due to low wind speed and boundary layer height leading to a lower resuspension of mineral dust in the ambient atmosphere. The Potassium (K) content, however, increased substantially owing to stubble/agriculture residue burning in the surrounding regions [as K is a tracer of biomass burning (Khare and Baruah, 2010)]. Similar results have been perceived in several other studies over Delhi (i.e., Jain et al., 2020; Rai et al., 2020). Therefore, this factor should be identified as crustal suspension and biomass burning. The second component with eigenvalue 5.21 represented 27.4% of the total variation with high loadings of S, Mn, Ni, Fe, and Pb. While Ni is related to vehicular emission especially with heavy diesel based vehicles (Khanna, 2015; Das et al., 2020), the elements such as Ni, Pb, Fe, and Mn are well associated with vehicular emissions mixed with road dust resuspension (Sternbeck et al., 2002; Khanna, 2015; Liu et al., 2015; Pant et al., 2015). The high concentration of S could be associated with coal combustion and secondary aerosols formation (Gummeneni et al., 2011). Therefore, this factor can be attributed to the vehicular emission, road dust resuspension, and secondary aerosols formation.

The third factor explains 20.6% of the overall variance with an eigenvalue of 3.9 and is strongly correlated with the elements Br, Cl, Cu, Rb, and Zr. These elements can have origin from industrial activities and solid-waste burning (Khodeir et al.,

2012). Br in previous studies has been identified as the element associated with industrial emissions probably from various drug and chemical manufacturing industries (Kothai et al., 2011). Halogens can also be produced from solid-waste burning. In Delhi, most of the incineration of dumped waste occurs in three locations viz, Okhla (also a major industrial area in South Delhi), Bhalswa (North Delhi) and Ghazipur (East Delhi) (Ghosh et al., 2019). Moreover, in post-monsoon and winter season, brick kilns have been reported to function in areas encompassing Delhi, and they can possibly elevate the Cl concentrations in the atmosphere. These brick kilns are operating in large numbers around Delhi-NCR (National Capital Region) due to heavy demand in the infrastructure sector (Rai et al., 2020). These traditional kilns use coal and biomass as fuel to bake bricks which could emit a large amount of Cl and Br (Rai et al., 2020). Cu, Rb and Zr are associated with industrial emissions from different electroplating and other alloy manufacturing industries (Hazarika et al., 2015). Chowdhury et al. (2017) in their study in Delhi have indicated major point sources of emissions from coal based thermal power plants as well as major industrial areas located in central, northern, and eastern part of Delhi. The total number of coal based thermal power plants in Delhi-NCR is 6. These power plants and numerous industrial areas emit large amount of PM and other pollutants (Mittal et al., 2012). Therefore, this factor can be represented as coal combustion, industrial emissions, and waste incineration.

CONCLUSIONS

The seasonal variation in mass concentration and elemental composition of PM2.5 was analyzed for the monsoon (June-July) and post-monsoon (October-November) seasons in 2019 at Delhi, India. The average PM25 concentrations for the monsoon and post-monsoon season were 42.2 \pm 15.5 μ g m⁻³ and 121.4 \pm 53.6 μ g m⁻³, respectively. High wind speed and low relative humidity (%) were observed in the June-July months of monsoon season as compared to those during the post-monsoon season. The elements that contributed most to the PM2.5 compositions were Si, Al, Na, and Mg in the monsoon season and S, Al, Na, Si, K and Cl in the post-monsoon season. Air-mass back trajectory analysis was performed to distinguish the major atmospheric pathways through which the air pollutants particularly PM2.5 are impacting at the receptor site. High transboundary contribution to PM_{2.5} was observed in the monsoon season, whereas in the post-monsoon season the contribution was by-and-large from the regional sources. PCA analyses, during the postmonsoon season, identified the major sources as (i) biomass burning and uplifted mineral dust, (ii) vehicular emissions, road dust resuspension, and secondary aerosols formation, and (iii) industrial emission, coal combustion, and solidwaste burning. These results were supported by the AIRS dust score and MODIS fire count data. The present study aims to assist the stakeholders and policymakers to better understand the characteristics of PM2.5 during the post-monsoon

season and to design and implement effective and efficient policy strategies to curb the problem of $\rm PM_{2.5}$ pollution in Delhi.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding author/s.

AUTHOR CONTRIBUTIONS

AM and VB have designed the work and drafted the manuscript. VB, MJ, and AM have collected the required data. Data analysis, data interpretation and final editing is done by AM, VB, MJ, and PR.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/frsc.2021. 648551/full#supplementary-material

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Assessment of Health Impact of PM_{2.5} Exposure by Using WRF-Chem Model in Kathmandu Valley, Nepal

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PM_{2.5} is one of the major air pollutants in Kathmandu Valley, and its emission and the unique atmospheric condition of the valley make it significantly hazardous to human health. The air pollution due to particulate matter is a major health issue with numerous negative impacts on us. This research aims to quantify the health impacts of PM25 exposure on the population of Kathmandu Valley. The ambient PM_{2.5} concentration of Kathmandu Valley was simulated using WRF-Chem model by using a horizontal grid resolution of 3 × 3 km. The concentration obtained from WRF-Chem was used as input in the health equation of an intervention model to quantify the health impacts. This quantitative assessment of atmospheric pollution was applied to evaluate the human exposure to PM_{2.5} in Kathmandu Valley. PM_{2.5} concentration and its distribution in the valley along with the ward-wise population distribution were used to find the health impact of the particulate matter in December 2019 in Kathmandu Valley. Exposure analysis using the model showed that 19 people could die due to lung cancer and 175 people could die due to all cause diseases except accidents due to PM_{2.5} exposure in December 2019. It was estimated that the reduction in the PM2.5 level by half in the valley reduces the monthly mortality by 51.4%. Hence, the exposure analysis of the particulate matter on the urban population could be improved by using air quality models in order to solve the health problems arising from air pollution.

Keywords: air quality, exposure, health impact model, Kathmandu Valley, WRF-Chem model, PM2.5

1. INTRODUCTION

 $PM_{2.5}$ in the ambient atmosphere is hazardous and detrimental to both the environment and human health. The amount of particulate matter can determine some of the impacts of air pollution on human health. If we consider the World Health Organization's recommended limits for the air pollution, the quality of Nepalese air is very poor with an annual mean $PM_{2.5}$ concentration of 44.5 $\mu g/m^3$, which exceeds the recommended threshold of 10 $\mu g/m^3$ (IQAir, 2019).

Studies have found that the major contributing factors of air pollution in the valley are the vehicles, residential combustion, brick kilns, the manufacturing and construction industries, biowaste burning, and dust particles (Stone et al., 2010; Kim et al., 2015; Sarkar et al., 2017; Shakya et al., 2017). Out of them, vehicular emission is the primary contributing factor due to consumption of fossil fuels (Sarkar et al., 2017; Shakya et al., 2017). The unusually high pollution levels from the vehicles are mainly due to the haphazard traffic system, poor maintenance of the vehicles, and ineffective control of vehicle emissions (CEN and ENPHO, 2003).

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Tuladhar A, Manandhar P and Shrestha KL (2021) Assessment of Health Impact of PM_{2.5} Exposure by Using WRF-Chem Model in Kathmandu Valley, Nepal. Front. Sustain. Cities 3:672428. doi: 10.3389/frsc.2021.672428 The Kathmandu Valley is the capital city of Nepal and has the largest number of population. One of the main reasons for the highly polluted state of Kathmandu Valley is the highly dense concentration of people coming from all over the country (Bakrania, 2015). Nepal has one of the highest rates of urbanization rates in the world that can steadily keep on increasing in the long run as well (Bakrania, 2015). Most of the development work has been concentrated in the Kathmandu Valley and the subsequent economic activities as well as the consumption of fossil fuel have aggravated the pollution and health-related problems in the city.

The traffic scenario in Kathmandu Valley is deteriorating steadily with the constant influx of people. A projection has estimated this increase in the number of vehicles in the valley from 461,927 in 2009 to 895,802 in 2021 (ADB, 2010). The increase in vehicular air pollution affects the human respiratory system. As the vehicular traffic grows, so does the production of $PM_{2.5}$ that results in adverse health impacts like respiratory and pulmonary diseases (US EPA, 2020). Ghimire and Shrestha (2014) showed that the total estimated $PM_{2.5}$ was 9646.40 tons/year from vehicular emission in Kathmandu Valley.

The InMAP model has been used to estimate the annual average primary and secondary PM2.5 related to changes in emissions. This model provides marginal health damages based on source-receptor relationships calculated by the WRF-Chem chemical transport model. Inputs to the model include precursor emissions (NH₃, SO₂, PM₂, NOx, and VOCs), annual average meteorology, air quality, deposition information, annual gridded surface emissions, and point sources. InMAP model uses population and health incidence data that can be used to estimate health impacts (IEc, 2019). EASIUR is another model that calculates the monetized health impacts of emissions changes in the contiguous United States. The EASIUR model follows a standard method for estimating mortality burden, which is an impact pathway analysis of estimating ambient PM2.5 from precursor emissions and then quantifying associated premature deaths based on epidemiological studies (Heo et al., 2016). This method achieves detailed spatial resolution according to the location of the emission source, accounting for differences in the exposed population downwind. EASIUR can quantify the benefits of the air quality policy scenarios and can be normalized per ton of emissions. Using chemical transport model simulations, the model builds a large dataset of air quality public health impacts from marginal emissions throughout the United States, taking into account the exposed population distribution throughout the country (Heo et al., 2016). APEEP model also follows a similar format of quantifying the health impacts. The model begins with calculating emissions using an air quality model. Next, APEEP computes exposures of people to ambient air pollution to determines the resulting human health impacts. APEEP calculates human exposures to the predicted concentrations by multiplying county-level pollution concentrations by the county-level population data. The model translates exposures into physical effects using concentrationresponse functions published in peer-reviewed studies in the epidemiological literature (Groosman et al., 2011).

The relationship between the change in concentration of the pollutant (Δx) , and the corresponding change in the population health response (Δy) derived from a concentration response relationship is the health impact function (US EPA, 2010). Many epidemiological studies report some measure of the change in the population health response associated with a specific change in the pollutant concentration, which is called the relative risk.

Pope et al. (2002) assessed the relationship between longterm exposure to fine particulate air pollution and all-cause, lung cancer, and cardiopulmonary mortality. The study found that each 10 μ g/m³ elevation in fine particulate air pollution was associated with 4, 6, and 8% increased risk of all-cause, cardiopulmonary, and lung cancer mortality, respectively. The findings are based on 16 years of research involving about 500,000 people and 116 metropolitan areas in the United States. Similarly another study by Krewski et al. (2009) consisted of approximately 360,000 participants residing in areas of the country that have adequate monitoring information on levels of PM2.5 analyzed the relative risks of all causes, cardiopulmonary disease (CPD), ischemic heart disease, lung cancer, and all remaining causes. The study showed 6% increase in overall mortality for every 10 μ g/m³ increase in PM2.5 concentration. Lepeule et al. (2012) found significant associations between PM_{2.5} exposure and increased risk of all-cause, cardiovascular and lung cancer mortality, and reported that each increase in $PM_{2.5}$ (10 µg/m³) was associated with an increased risk of all-cause mortality of 14%, along with 26 and 37% increase in cardiovascular and lung cancer mortality, respectively.

The health impact of $PM_{2.5}$ exposure in Nepal has been conducted using concentration–response relations from various other regions (Gurung and Bell, 2013). Giri et al. (2007) have estimated the mortality due to the PM_{10} pollution in Kathmandu Valley as 17,132 premature deaths/year. Shah and Nagpal (1997) estimated the impact of PM on mortality and morbidity and found the impact of 84 deaths, 506 cases of chronic bronchitis, 475,298 restricted activity days, and 1.5 million respiratory symptom days in Kathmandu Valley in 1990. In this study, the $PM_{2.5}$ pollution exposure in Kathmandu Valley and its health impacts have been evaluated using the distributed approach of simulating the air quality by employing WRF-Chem modeling.

The annual $PM_{2.5}$ concentration of Nepal is 10 times the limit set by WHO. The strongest predictor for deaths caused by ambient air pollution is chronic $PM_{2.5}$ exposure (Künzli et al., 2001; Pope and Dockery, 2006). $PM_{2.5}$ is capable of traveling long distances and can also be highly spatially variable near emission sources. To date, limited studies have been conducted to analyze the $PM_{2.5}$ emission and its health impacts in Kathmandu Valley. This study aims to use a high-resolution WRF-Chem simulation to test and support the research on $PM_{2.5}$ emission and its health impact.

2. MATERIALS AND METHODS

2.1. Study Area

The research was conducted for Kathmandu Valley as shown in **Figure 1**, having a population of 1,376,000 in 2019, and 665 km² area (Macrotrends, 2020). The valley stands at 1,425 m (4,675



ft) above sea level. It comprises three main districts of Nepal: Kathmandu, Lalitpur, and Bhaktapur District. The total number of wards in each district are 71 wards in Lalitpur, 38 in Bhaktapur, 138 in Kathmandu. The temperature in Kathmandu valley is around $20-35^{\circ}$ C in the summer and -3 to 20° C in winter.

2.2. Study Period

In order to select the time period for the study, US embassy station data were analyzed. Upon analyzing the PM concentrations throughout the year of 2019, we observed that PM peaks in the driest winter month of December. In December, the average temperature ranges between min 3.7°C (38.7°F) and max 20.2°C (68.4°F), during which PM concentration is the maximum (IQAir, 2019; Weather Atlas, 2020). The pollution stations of Government of Nepal started operating only from 2016, and US Embassy stations operated after a year in 2017. During this research, only 3-year data were available but the data were very limited with lots of missing data. In a comparison of monthly data, we found that December, January, and February usually have high PM concentrations. For example, in US embassy station data of 2018, December had the highest PM_{2.5} concentration of 101.6 μ g/m³ followed by January and February with 99.9 and 94.8 µg/m³, respectively. The PM_{2.5} exposure analysis of this study was done in very high spatial resolution of 3×3 km. Some papers (Heo et al., 2016; Thind et al., 2019) have used 36 \times 36 km and 48 \times 48 km resolution. Being a high-resolution simulation with a time period representing the highest PM2.5 pollution in Kathmandu Valley, the time period from December 1, 2019 to December 30, 2019 was selected for the study.

2.3. Population Data

The baseline population data from Central Bureau of Statistics (CBS), Nepal (https://cbs.gov.np/population/) was used for the study. Using the Ratio method as used by CBS reports for population projection, this ward-wise population data were projected from the baseline year 2011 to year 2019.

The equation of this method is:

Sub area Projected Population =
$$\frac{Sub area Population}{Parent Population} \times Parent Population Projection$$

where,

Sub area Projected Population is the required ward-wise projected population for 2019,

Sub area Population is the ward-wise population of baseline year 2011,

Parent Population is the Population of Nepal in baseline year 2011,

Parent Population Projection is the projected population of Nepal in 2019.

The *Parent Population* was taken from the Population Projection Report prepared by CBS (2014).

2.4. Mortality Data

The mortality rate for lung cancer and all-cause mortality data to quantify the health impacts were taken from the reports published by Nepal Health Research Council (NHRC). The all-cause mortality rate for the valley is found to be 607.8 per 100,000 people (NHRC, 2018).

TABLE 1 | WRF-WPS model setup.

WRF-Chem parameters	Value
Horizontal grid resolution	3 km
Grid	66×58 cells
Vertical resolution	38 levels
Projection	Mercator

TABLE 2 | Relative risk for the diseases from epidemiological study done by Pope et al. (2002).

Cause of mortality	Relative risk (R)
Lung cancer	1.14
All cause	1.06

2.5. WRF-Chem Modeling

The Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) is a widely used distributed model to simulate the atmospheric process and it is able to model meteorological processes and atmospheric transport.

WRF-Chem is a numerical weather and atmospheric simulation model that is coupled with chemistry (Grell et al., 2005). It can simulate and predict the atmospheric emission of pollutants, the chemical transport, and transformation at multiple spatial scales. The interaction between meteorology and chemical processes is also modeled by WRF-Chem.

2.6. Modeling Setup

Version 4.0 of Weather Research and Forecasting Model coupled with Chemistry was used for simulating the meteorology and chemistry over the model domain shown in **Table 1**. The model domain is defined on the mercator projection. The model contains 3 km grid resolution and of 38 vertical layers.

The data required for the model run were downloaded from different sources.

NCEP FNL Operational Global Analysis was used for the initial and lateral boundary conditions for meteorological field. Fire Inventory from the National Centre for Atmospheric Research (NCAR) (FINN) version 1.5 was used for biomass burning data. The anthropogenic emission required for running the model is taken from Emission Database for Global Atmospheric Research (EDGAR-HTAP v2) inventory. The biogenic emissions were from Model of Gases and Aerosols from Nature (MEGAN). WSM 5-class microphysics scheme, Grell 3-D cumulus parameterization, RRTM longwave option, Dudhia shortwave scheme, Yonsei University planetary boundary layer parameterization, MOZCART chemistry scheme, and Madronich F-TUV photolysis option were used for running the WRF-Chem model.

2.7. Estimation of PM_{2.5} Pollution-Related Deaths

Health impact function was used to estimate the number of deaths and illnesses associated with $PM_{2.5}$ attributable to each of the 247 wards of Kathmandu Valley. One of the input parameters is the $PM_{2.5}$ concentration from the WRF-Chem model.

Various reduced complexity models (RCMs) have been developed to quantify and value the health impacts of changes in air quality. The reduced form models employ simpler methods to approximate the more complex analyses with a lower computational burden (IEc, 2019). Health benefit results from the models can be obtained by the use of Chemical

Transport Models (CTMs) to estimate the impact of emissions on ambient concentrations, and the health effects from exposure to these concentrations can be quantified by using concentration– response functions (Gilmore et al., 2019). Several RCMs such as EASIUR (Heo et al., 2016), InMAP (Tessum et al., 2017), and APEEP (Muller and Mendelsohn, 2007) use health impact function from BenMAP (US EPA, 2010) to determine the mortality due to pollutant exposure. The function calculates the number of death resulted per year due to PM exposure by taking a certain concentration response relation (Pope et al., 2002; Krewski et al., 2009). All three RCMs use the same equation to predict the mortality burden caused by PM_{2.5} pollution.

The log-linear relationship is the most common form of health impact function used in these types of studies (US EPA, 2010). A typical health impact function specifying a log-linear relationship between risk and air quality change is as follows (US EPA, 2010):

$$y_i = \left\{ \exp\left(\frac{\log R}{10} \cdot c\right) - 1 \right\} \times Total Population \times Mortality Rate (1)$$

where

c is the average PM_{2.5} concentration,

Total Population is the ward-wise population,

Mortality Rate is the mortality rate for certain diseases per 100,000 people,

R is the relative risk reported from various published epidemiological studies,

 y_i is the estimated number of PM_{2.5}-related total deaths for each ward.

In this study, the value of *R* is different for lung cancer and allcause mortality (**Table 2**). Since Nepal does not have a reliable published source of relative risk, we have taken it from the study conducted by Pope et al. (2002), which states that an increase of 10 μ g/m³ in fine PM concentration in the atmosphere contributes about 6 and 14% increase in the risk of all cause and lung cancer mortality, respectively. Due to lack of mortality data for Kathmandu Valley, only two factors—lung cancer and all cause disease—could be considered.

The input and output of the model were plotted in the Kathmandu Valley map by using the QGIS software.

3. RESULTS

3.1. Projected Population

The ward-wise population of Kathmandu Valley for the year 2019 was estimated using the Ratio method. The baseline population was taken from Central Bureau of Statistics (2011). **Figure 2** shows the projected population of the valley for 2019.





Kathmandu Valley at 2019 has approximately 2.7 million people. The densely populated urban centers comprise the Kathmandu and Patan cities and Bhaktapur city in the eastern edge of the valley. Urbanization can be seen increasing throughout the valley, agricultural field are also present over the flat valley floor, and substantial area is found to be used for brickfields especially in the Bhaktapur district.

3.2. Mortality Rate

Figure 3 shows the mortality rate of Kathmandu Valley for lung cancer (NHRC, 2018). Mortality rate (per 100,000 people) due to all cause (except accidents) diseases in Kathmandu valley was taken as 607.8 (NHRC, 2018). We projected the rate from 2018, using Ratio method, to 2019 but no significant changes were observed. So we used mortality rate of 2018.



3.3. Emission Concentration From WRF-Chem

In **Figure 4**, the 1 month (December, 2019) average spatial distribution of calculated $PM_{2.5}$ is shown along with the population distribution in the Kathmandu valley. The maximum $PM_{2.5}$ concentration is seen in Kathmandu district. This region of Kathmandu, being the main urban center, holds the maximum population density along with maximum traffic congestion, with large development activities resulting in high air pollution. Largely protected from any winds, the pollutants are not dispersed and hang heavy over the local population especially during the dry winter months. Wards including Nagarjun 4, Nagarjun 9, Nagarjun 10, Chandragiri 13, and Chandragiri 14 experience the greatest $PM_{2.5}$ concentrations. The lowest concentration was found to be at Bagmati 4. There is less $PM_{2.5}$ concentration in the outskirts of Kathmandu, lower region of Lalitpur and Bhaktapur district.

3.3.1. Health Impact of PM_{2.5} Exposure

The cumulative deaths due to PM_{2.5} induced lung cancer and all cause is found to be 19 and 175, respectively, for December 2019, which was obtained using Equation (2) that estimates the number of PM_{2.5}-related total deaths (*Y*) for each ward *i* (i = 1, 2, 3... 247).

$$Y = \sum_{i=1}^{247} y_i$$
 (2)

where y_i is the ward-wise PM_{2.5}-related death (see Equation 1) and Y is the total PM_{2.5}-related death of entire Kathmandu Valley.

The health impact function was applied to the 247 wards individually to calculate the mortality burden per ward. The summation of all the ward results provided us with the total number of deaths in the entire valley. **Table 3** shows that the death number for lung cancer and all cause disease are 19 and 175, respectively. Different relative risks were applied for lung cancer and all cause, along with different mortality data input to provide the mortality results.

TABLE 3 | PM_{2.5} related premature deaths attributable to emissions in December 2019.

	Total no. of death	Average mortality rate (per 100,000)
Lung cancer	19	0.71
All cause	175	6.37

4. DISCUSSION

Figure 5 shows maximum and minimum mortality values for lung cancer to be 0.403 and 1.081 per 100,000 people, respectively. The highest $PM_{2.5}$ induced lung cancer mortality rates are seen in Lalitpur district. The mortality rate for lung cancer is 41.6 (per 100,000) as provided by NHRC (2018), which explains the higher $PM_{2.5}$ induced lung cancer mortality rates than the other two districts. Although the population density of Kathmandu is greater, the lung cancer mortality rate for Lalitpur district is greater than that of both Kathmandu and Bhaktapur districts. The topmost region of Lalitpur district has the maximum mortality rates due to high emission concentration as well as large population.

The maximum and minimum mortality rate burden for all cause is shown in **Figure 6** to be 4.31 and 6.93 per 100,000 people, respectively. The highest $PM_{2.5}$ induced all cause mortality rates are seen in Kathmandu district. The mortality rate for all cause is 607.8 (per 100,000) for the entire valley. It is evident that the high emission concentration and population cause maximum mortality burden due to $PM_{2.5}$ in Kathmandu district. Therefore, $PM_{2.5}$ -related mortality burden is highest where population concentration and $PM_{2.5}$ emission is maximum.

The estimated mortality is higher than that reported by Regmi and Kitada (2003). Regmi and Kitada (2003) estimated 226 deaths per year, whereas this study estimates around 175 deaths during the month of December (**Table 4**). Increase of both the population and pollutant concentration has contributed toward this trend. The health impact functions (Ostro and Chestnut, 1998) used by Regmi and Kitada (2003) provides very less relative risk compared to ours, and the target population for the valley was also 50% less than the target population of this study. This change in the health impact constant and the population distribution predicted lesser mortality results.

Similar study was done by Shah and Nagpal (1997), who estimated similar results as that of Regmi and Kitada (2003). Shah and Nagpal (1997) had also used the same health impact equation for similar target population as that of Regmi and Kitada (2003). The main reasons for the difference in the results are the increases in population and pollutant concentration.

The mortality results due to $PM_{2.5}$ emission was calculated after reducing the concentration of every ward by half. The hourly $PM_{2.5}$ concentration data are not compared with the observed station data since the model provides average of the grid (3 km) while the stations provide point wise data. The grid value and the station point data are not comparable. The monthly data obtained for the individual wards got reduced by nearly 50% and





the results were obtained (**Table 5**). Upon analyzing the result before and after the emission reduction, it is seen that the death result also significantly reduced by half. Thus, it is estimated that reduction in $PM_{2.5}$ level in the valley will reduce the monthly mortality by approximately 50%.

Taking into account the exposure map, high concentration of exposure seems to be prevalent in the cities of Kathmandu district. To reduce adverse health outcome stringent emission control measures should be implemented (Regmi and Kitada, 2003). Nepal is regularly ranked poorly in the air quality

PM_{2.5}'s Health Impact in Kathmandu

Death	Time period
175	December 2019
226	Annual 2003
84	Annual 1996
	175 226

TABLE 5 | Comparison between $PM_{2.5}$ resulted death after reducing the emission by half in December 2019.

	PM _{2.5} resulted death	PM _{2.5} resulted death after reducing emission	Percentage decreased (%)
Lung cancer	19	9	52.6
All cause	175	85	51.4

indices. Considering the mortality burden, the places affected by PM_{2.5} pollution should be identified and effective air pollution mitigation plan should be enforced.

4.1. Conclusion and Recommendations

In this research, numerical simulation of particulate matter of Kathmandu Valley was done using Chemical Transport Model, WRF-Chem for December, 2019. Values of simulated 24-h mean PM_{2.5} concentration ranges from 30 μ to 65 μ g/m³. The values of PM_{2.5} obtained from the model simulation exceeds the WHO standard, which is 25 μ g/m³ 24-h mean. The level of PM_{2.5} in Kathmandu's air is extremely high, especially in the dry winter month (December). In this month, the air in urban Kathmandu can be classified as "very unhealthy."

Human $PM_{2.5}$ pollution exposure has been studied utilizing the numerically simulated $PM_{2.5}$ concentration value and residential distribution of population over Kathmandu valley. Human- $PM_{2.5}$ pollution-exposure maps for Kathmandu valley have been analyzed and various health outcomes due to the prevalence of $PM_{2.5}$ are evaluated. Spatial exposure strength, i.e., population times concentration for $PM_{2.5}$, occurs in the main urban area of Kathmandu, Patan, Thimi, and Bhaktapur cities due to the large population living in these cities. From the model, we calculated the total number of deaths for the month of December, 2019 to be 19 from lung cancer and 175

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from all cause disease, both $PM_{2.5}$ induced. The results obtained showed that greater the emission and population concentration, greater is the mortality rate in the region. From our calculations, mortality burden was highest in central Kathmandu with a value of 1.081 and 6.93 per 100,000 people for lung cancer and all cause diseases, respectively. The highest $PM_{2.5}$ concentration is seen in the central region of Kathmandu Valley where the population density is also high, thereby contributing to highest mortality in that area. If the monthly average emissions are reduced by 50%, the total number of deaths are also approximately reduced by 50%, implying that $PM_{2.5}$ is directly related to total number of deaths in this case. Finally, the results indicate that $PM_{2.5}$ is dangerous to human health and can even be fatal.

This research plays a role in understanding the health impacts caused by $PM_{2.5}$ based on quantitative analysis. Since annual time period was not possible for this study, a future study with a longer time period could be proposed. Other air pollutants can also be studied in the future along with their health impacts. Moreover, study including whole Nepal as study area can also be proposed to further understand the relation between $PM_{2.5}$ and its human health impact. There are evidences that link air pollution and health impacts and further studies are required on the health effects of air pollution in Kathmandu valley. The result from these studies can be used by policymakers to improve the air quality in Nepal. We also recommend the simulation for a longer time period at high resolution to better understand the relation between $PM_{2.5}$ concentration and number of death attributable to it.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

PM carried out the GIS processing of input and output data in the exposure maps. AT performed the population projection analysis. KS performed the simulations of WRF-Chem and coordinated the entire research. All authors contributed to the article and approved the submitted version.

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Long-Term (2003–2019) Air Quality, Climate Variables, and Human Health Consequences in Dhaka, Bangladesh

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Pavel MRS, Zaman SU, Jeba F, Islam MS and Salam A (2021) Long-Term (2003–2019) Air Quality, Climate Variables, and Human Health Consequences in Dhaka, Bangladesh. Front. Sustain. Cities 3:681759. doi: 10.3389/frsc.2021.681759 Long-term trends in air quality by studying the criteria pollutants (PM_{2.5}, PM₁₀, CO, O₃, NO₂, and SO₂) and climate variables (temperature, surface pressure, and relative humidity) were depicted in this study. The 17-year (2003–2019) average values of PM_{2.5}, PM_{10} , CO, O₃, NO₂, and SO₂ were 88.69 \pm 9.76 μ g/m³, 124.57 \pm 12.75 μ g/m³, 0.69 \pm 0.06 ppm, 51.42 \pm 1.82 ppb, 14.87 \pm 2.45 ppb, and 8.76 \pm 2.07 ppb, respectively. The trends among the ambient pollutants were increasingly significant (p < 0.05) except for O₃ with slopes of 1.83 \pm 0.15 μ g/m³/year, 2.35 \pm 0.24 μ g/m³/year, 0.01 \pm 0.002 ppm/year, 0.47 \pm 0.03 ppb/year, and 0.40 \pm 0.02 ppb/year for PM_{2.5}, PM₁₀, CO, NO₂, and SO₂, respectively. Pearson correlations revealed a significant association among the pollutants while a noteworthy correlation was observed between ambient pollutants and surface temperature. Principal component analysis (PCA) and positive matrix factorization (PMF) have been employed collectively to examine the main sources of the pollutants. PCA revealed similar trends for PMs and CO, as well as NO₂ and SO₂ being equally distributed variables. PMF receptor modeling resulted in attributing four sources to the pollutants. The factors inferred from the PMF modeling were signified as vehicular emissions, road/soil dust, biomass burning, and industrial emissions. The hazard quotient (HQ) values were not antagonistic (HQ < 1) in acute exposure levels for the three age groups (infants, children, and adults) while showing significant health risk (HQ > 1) in chronic exposure for infants and children. Children are identified as the worst sufferers among the age groups, which points to low breathing levels and high exposure to traffic pollution in Dhaka, Bangladesh.

Keywords: air quality, climate variables, positive matrix factorization, hazard quotient, Kendall and Spearman's correlations, Pearson correlations, principal component analysis

HIGHLIGHTS

- Long-term trends of criteria air pollutants and climate variables were analyzed.
 - Significant positive trends were observed for the pollutants except ozone.
- Four factors were characterized as estimated sources from PMF modeling.
- HQ exceeded the acceptable limit (>1) for chronic exposure to children and infants.



INTRODUCTION

Air pollution and climate change are the two most significant and interconnected challenges plaguing the 21st century. If existing policies remain unchanged, air pollution would be the most important environmental factor affecting premature deaths by 2050 (OECD, 2012). Global warming has culminated in a 43% increase in anthropogenic radiative forcing (RF) since 2005, according to the new IPCC report, focusing on human behavior as the most significant influencer (Stocker et al., 2013). The urban heat-island effect causes towns to be far warmer than their neighboring rural area due to enhanced anthropogenic activities. Owing to associations between warming and air emissions, this effect would pose extreme health problems for cities around the world by 2050 (Schmale et al., 2014). Lancet Commission on Pollution and Health reported 6.5 million deaths in 2015 due to atmospheric air pollution (Landrigan et al., 2017). while mortality due to past climate change on air quality was much lower: \sim 1500 and \sim 2200 deaths per year due to ozone and PM_{2.5}, respectively (Silva et al., 2013). The global air quality scenarios are unlikely to change anytime soon, and the crisis in the megacities of developing countries is expected to worsen. As a result, it is important to continue tackling air quality problems in tandem with climate change mitigation initiatives (Williams, 2012).

Approximately 91% of the world's population live in areas where WHO air quality standards have been exceeded. However, due to its large population, Bangladesh is one of the most polluted countries in terms of air quality, with Dhaka ranking as one of the world's two most polluted cities (AirVisual, 2018). In Bangladesh, there had been little to no effort and administrative works to track or mitigate ambient air pollution before 1999. In 1999, the government began establishing frameworks and regulations to meet US EPA and Bangladesh National Air Quality Standards, especially in Dhaka. In this regard, a number of controls have been implemented, including the prohibition of leaded gasoline in July 1999 and the replacement of old two-stroke engine three-wheelers with compressed natural gas (CNG)-powered four-stroke three-wheelers beginning in January 2003; introducing CNG-powered cars, buses, and trucks; and regulating brick kiln emanations, which resulted in a reduction of airborne Pb concentrations and improved air quality than before (Salam et al., 2013; Begum and Hopke, 2019). However, since the introduction of CNG and the prohibition of two-stroke engines, traffic congestion in Dhaka city has greatly worsened air quality, and numerous brick kilns using crude oil, fossil fuel, coal, natural gas, electricity, and biomass as a source of energy have sprung up all over the city and have been discharging many air pollutants including black carbon and organic carbon (Salam et al., 2013). In addition, research on the results of CNG conversion in other megacities (Rio de Janeiro, Mexico City, and New Delhi) and the greenhouse gas (GHG) benefits of such conversion showed that switching from diesel vehicles increased high-emission particulates and black carbon, which are more potent as GHGs than CO₂ or CH₄. Thus, large-scale conversion of petrol vehicles embodies the risk of reduced GHG benefit or even negative GHG impacts (Wadud and Khan, 2013). Furthermore, permanent wetlands have been disappearing at an unprecedented pace as a result of unplanned urbanization to accommodate the huge population, with more than 49% of wetland areas disappearing in Dhaka city between 1960 and 2008 (Rai et al., 2017). As a result of such adverse situations, Dhaka had very high (>1) toxicity potential (TP) values of $PM_{2.5}$ and PM_{10} (Zaman et al., 2021) and recorded the highest mortality and morbidity rates (hospital admissions) among the megacities studied, with about 7,000 deaths and 2,100 excess cases (cardiovascular and respiratory) each year (Gurjar et al., 2010). **Table 1** reports several previous researches on ambient air quality and climate impact in Dhaka, Bangladesh, undertaken after 2003.

Carbon monoxide, lead, ground-level ozone, particulate matter (PM), nitrogen dioxide, and sulfur dioxide have been designated as criteria air contaminants by the Environmental Protection Agency (EPA) and the World Health Organization (WHO), and national and global ambient air quality standards (NAAQS) have been developed for these six pollutants (EPA). These contaminants, which are made up of various materials, have a broad range of sources of pollution and varying degrees of health impacts and toxicity, dictating the use of source apportionment studies to understand the formation processes and their sources. On source apportionment, two chemometric techniques, principal component analysis (PCA) and positive matrix factorization (PMF) receptor model, were employed collectively. PCA is utilized to relate the air pollutants and their overall changes in the aerosol composition while PMF is used to determine the different sources of pollution and the temporal variability of each pollutant without considering their correlations (Padoan et al., 2020).

Dhaka is one of the most contaminated cities in the world and is in dire need of effective mitigation policies to attain national and global air quality standards. Unfortunately, there is limited information for the criteria air pollutants levels, sources apportionment, impact on human health, and climate variables based on long-term datasets. Therefore, the objective of this study is to understand the long-term (2003-2019) evolution of major atmospheric contaminants (PM2.5, PM10, CO, O3, NO2, and SO_2) and the effect of meteorological parameters (temperature, pressure, and relative humidity) on these pollutants following the January 2003 ban on old two-stroke engine three-wheelers in favor of CNG-powered four-stroke three-wheelers. Although there have been recent comparative studies for fine and coarse fractions of PM in terms of source attribution in the seasonal and short-term periods, long-term studies for all of the criteria contaminants and their health effects, as well as their origins, have been scarce. This study will help evaluate the aftermath of implementing the CNG-powered vehicles and how the vehicles and other sources might be regulated in the future to improve the ambient air quality in Dhaka, Bangladesh.

Description of the Study Area

Dhaka, Bangladesh's capital and most populous city, is situated at the northeastern end of the Indo-Gangetic Plain (IGP). Dhaka is the center of Bangladesh with an elevation of \sim 9 m above sea level (**Figure 1**). Dhaka encompasses a total area of 1463.60 square kilometers (565.10 square miles) and possesses a population of 12.5 million and, hence, is one of the most densely populated cities in the world with a density of 8,229 per square kilometer. Dhaka has a population growth rate of 3.48% per year due to ever-growing urbanization (urbanization rate = 77.36%), which drives a huge chunk of the population toward urban life every year from rural/suburban areas (www.dhaka.gov.bd). As

Studied variables	Observation	Model/study methods	References
PM _{2.5} , its precursors, and climate variables	Air quality changes from the CNG conversion scheme in 2010 resulted in around 2045 (1665) avoided premature deaths and a savings of around USD 400 million in greater Dhaka. Climate expenses (~USD 17.7 million) were in the order of magnitude less than the air quality gains.	GIS-based modeling	Wadud and Khan, 2013
Climate variables (monthly rainfall and temperature)	Daily energy usage in Dhaka city rises by 6.46–11.97 and 2.37–6.25 MkWh per unit increase in temperature and rainfall, respectively, while daily gross residential energy demand and peak demand will rise by 5.9–15.6% and 5.1–16.7% by the end of the century, depending on climate change scenarios.	An ensemble of six GCMs of CMIP5 under four RCP scenarios	Shourav et al., 2018
PM_{10} , $PM_{2.5}$, BC, and Pb	Dhaka's air quality has remained steady over the last decade, despite increased economic development and the number of sources such as passenger cars and brick kilns.	EEL-type Smoke Stain Reflectometer	Begum and Hopke, 2018
PM _{2.5}	Wood burning, soil dust, brick kilns, fugitive Pb, road dust, Zn sources, motor vehicles, and sea salt have been reported as sources.	lon Beam Analysis (IBA) and EEL-type Smoke Stain Reflectometer	Begum and Hopke, 2019
NO ₂ , CO, O ₃ , SO ₂ , and PM _{2.5} and PM ₁₀	The presence of a seasonal pattern of air quality implies that the air is highly toxic and polluted.	Seasonal Autoregressive Integrated Moving Average (SARIMA) model	Islam et al., 2020
PM _{2.5}	Metal(oid) contamination in dust and soil at school compounds was primarily caused by traffic-related events, natural causes, and manufacturing operations.	Ion Beam Analysis (IBA) methods and PCA-APCS-MLR receptor model	Rahman et al., 2021

GCM, global circulation models; CMIP5, coupled model intercomparison project phase 5; RCP, representative concentration pathway.



a result, Bangladesh with a mean $PM_{2.5}$ concentration of 77.1 μ g/m³ and megacity Dhaka with the same mean concentration of that of Bangladesh has emerged as the 1st and 2nd most polluted country and capital city in the world (Amato et al., 2020).

Air Pollution and Meteorological Data

Although the Department of Environment (DoE) has some ground-based monitoring stations for atmospheric pollutants, long-term datasets are still scarce for Dhaka. As a result, 24-h ambient concentrations of the air pollutants ($PM_{2.5}$, PM_{10} , CO, O₃, NO₂, and SO₂) and meteorological data (temperature, surface pressure, and relative humidity) were collected from the EAC4 (ECMWF Atmospheric Composition Reanalysis 4) global reanalysis dataset produced by the European Centre for Medium-Range Weather Forecasts (ECMWF) from Copernicus

Atmosphere Monitoring Service (CAMS) for the period January 2003 to December 2019. The dataset contains gridded global data with $0.75^{\circ} \times 0.75^{\circ}$ horizontal resolution and has the following vertical coverage: surface, total column, model levels, and pressure levels. This period was chosen to depict long-term air quality and its consequences on climate variables and human health in Dhaka, Bangladesh.

CAMS Reanalysis Data

Reanalysis combines model data with measurements from around the world to construct a globally robust and coherent resource using a physics and chemistry-based model of the atmosphere. The data integration is defined as a technique used by numerical weather prediction centers and air quality forecasting centers, in which a preceding prediction is merged with currently available measurements in an optimized way every so many hours (12h at ECMWF) to create a new best estimate of the atmospheric situation, known as analysis, from which an updated, enhanced forecast is produced (ads.atmosphere.copernicus.eu). The CAMS reanalysis is the most recent global reanalysis data collection of atmospheric composition (AC) developed by the Copernicus Atmosphere Monitoring Service, and it consists of three-dimensional time-consistent AC fields, including aerosols, chemical species, and greenhouse gases, through the CAMS global greenhouse gas reanalysis (EGG4). The dataset improves on the knowledge obtained during the development of the Monitoring Atmospheric Composition and Climate (MACC) reanalysis and the CAMS interim reanalysis. The assimilation method estimates observational biases and separates high-quality data from lowquality data. Estimates are made using the atmosphere model in places where data collection is limited or for pollutants for which no direct measurements are possible. Reanalysis is a very easy and common dataset to work with since it provides forecasts at each grid point across the world at each daily production time, over a long period of time, and in the same format.

The CAMS reanalysis was created with 60 hybrid sigma/pressure (model) levels in the vertical, with the top level at 0.1 hPa, using 4DVar data assimilation in ECMWF's Integrated Forecast System (IFS) CY42R1. The 4DVar data assimilation employs 12-h assimilation periods spanning 09 UTC to 21 UTC and 21 UTC to 09 UTC, respectively. The IFS model documentation for various model cycles can be found at https://www.ecmwf.int/en/forecasts/documentation-and-support/changes-ecmwf-model/ifs-documentation. On these levels, atmospheric data are available, which are also interpolated to 25 strain, 10 potential temperature, and 1 potential vorticity level(s). Data on the "surface" or "single stage" are also available.

EAC4 is the fourth-generation ECMWF global reanalysis of AC. Monthly surface data were utilized to study human exposure to the selected pollutants and the effect of the climate variables. Monthly means for assessments and instantaneous predictions are calculated using data with a valid time in the month, ranging from 00 to 23 UTC, except the time 00 UTC on the first day of the next month. Data with a prediction date that falls within the month are used to establish monthly means for accumulations and mean rates.

However, there have been some issues associated with assimilation of the CAMS global reanalysis (EAC4). During 2003 and between March and August 2004, the ozone analysis has a degraded quality, while from 2013 onwards, there is a larger seasonally varying bias in ozone in the free troposphere, particularly in the Arctic and Antarctic that is not seen in the control run. Furthermore, NO₂ in the CAMS reanalysis is mainly influenced by prescribed emissions (e.g., anthropogenic MACCity, GFAS biomass burning) and only to a lesser extent by assimilated findings due to its short lifespan. As a result, patterns or deviations derived from NO₂ reanalysis fields will primarily represent trends in underlying pollution. More details about the EAC4 data products, i.e., the observation techniques, data assimilation methods, bias correction, etc. have been described by Inness et al. (2019) and Flemming et al. (2015). **TABLE 2** | Observations used in the assimilation and validation of CAMS, ordered by species.

Species, Vertical range Assimilation Validation Aerosol mass (PM10, MODIS Aqua/Terra PM2.5) European Airbase stations O3, PBL/surface Surface ozone: WMO/GAW, NOAA/ESRL-GMD, AIRBASE CO, PBL/surface IASI, MOPITT Surface CO: WMO/GAW, NOAA/ESRL NO2, troposphere OMI, GOME-2, partially constrained due to short lifetime TROPOMI, SCIAMACHY, GOME-2, MAX-DOAS, TROPOMI SO2 GOME-2 (Volcanic eruptions) TROPOMI					
PM _{2.5}) Surface ozone: WMO/GAW, NOAA/ESRL-GMD, AIRBASE CO, PBL/surface IASI, MOPITT Surface CO: WMO/GAW, NOAA/ESRL NO ₂ , troposphere OMI, GOME-2, partially short lifetime TROPOMI, SCIAMACHY, GOME-2, MAX-DOAS, TROPOMI SO ₂ GOME-2 (Volcanic		Assimilation	Validation		
CO, PBL/surface IASI, MOPITT Surface CO: WMO/GAW, NOAA/ESRL NO ₂ , troposphere OMI, GOME-2, partially constrained due to short lifetime TROPOMI SO ₂ GOME-2 (Volcanic	(10)	MODIS Aqua/Terra	European Airbase stations		
NO ₂ , troposphere OMI, GOME-2, partially TROPOMI, SCIAMACHY, Constrained due to short lifetime TROPOMI SO ₂ GOME-2 (Volcanic	O ₃ , PBL/surface		,		
constrained due to short lifetime GOME-2, MAX-DOAS, TROPOMI SO2 GOME-2 (Volcanic	CO, PBL/surface	IASI, MOPITT	,		
-	NO_2 , troposphere	constrained due to	GOME-2, MAX-DOAS,		
	SO ₂	`			

MODIS, Moderate Resolution Imaging Spectroradiometer; IASI, Infrared Atmospheric Sounding Interferometer; MOPITT, Measurements of Pollution in the Troposphere; WMO/GAW, World Meteorological Organization/Global Atmosphere Watch; NOAA/ESRL-GMD, National Oceanic and Atmospheric Administration/Earth System Research Laboratory-Ground-based midcourse defense; GOME-2, Global Ozone Monitoring Experiment-2; TROPOMI, Tropospheric Monitoring Instrument; SCIAMACHY, Scanning Imaging Absorption Spectrometer for Atmospheric Cartography; MAX-DOAS, Multi-Axis Differential Optical Absorption Spectroscopy.

Validation of the Reanalysis Data

The CAMS-84 is a sub-project of CAMS that deals with service product validation. The validation reports for the global and regional services are updated every 3 months by CAMS-84. The validation is focused on a variety of measurements and measuring methods, including *in situ* observations, surface remote sensing, airplane observations, balloon sounding, ship observations, and satellite observations. With a deadline of \sim 1 month after sensing, the validation reports' three-monthly interval adds restrictions on the timely availability of the findings. **Table 2** represents the assimilation and validation products for specific species along with their vertical range. The validation reports and the verification websites can be found at http://atmosphere.copernicus.eu/user-support/validation/verification-global-services.

For this study, the selected reanalysis products were validated with the monthly average air pollution concentration data overlapping the months of the analysis period obtained from the Department of the Environment (DoE, CASE project) in Dhaka, Bangladesh. Validation of $PM_{2.5}$ and PM_{10} NO₂, and SO₂ with the DoE data has been depicted in the following figure and the concentrations showed R^2 values of 0.93, 0.85, 0.58, and 0.76, which confirms that the reanalysis data are capable of reflecting the ground-based air pollution data (**Figure 2**). Validation for the other pollutants was not done due to temporal anomalies between the reanalysis and ground-based data.

Source Apportionment Using PCA and PMF Modeling

Correlations among the variables were studied through Pearson correlation coefficients instead of non-parametric tests as nonparametric tests are better suited for datasets having no linear relationships or violating normality (Hauke and Kossowski,



2011). Source apportionment has always been tricky in the case of environmental datasets (Gao et al., 2016). PCA has been used robustly (Cotta et al., 2020) and coupled with other techniques (Sun and Sun, 2017; Franceschi et al., 2018; Gao et al., 2020; Liu et al., 2020). On the other hand, the contribution of various sources to PM and other contaminants around the world have been studied by PMF analysis, as well as references therein (Cesari et al., 2016, 2018; Sharma et al., 2016a,b; Crilley et al., 2017; Liu et al., 2017; Ryou et al., 2018; Jain et al., 2019, 2020). Although PCA is useful for evaluating the association between variables and measurements, it can also be used to describe each observation independently, whereas PMF is more based on the overall dataset definition. As a result, PCA can be used to explain how each attribute affects the observations, as well as whether certain observations or classes of observations have unusual characteristics. Instead of focusing on each observation or indicator, PMF explores the temporal pattern of all variables to find a potential common source. As a result, combining PCA and PMF to achieve an aggregate statistical summary of data is a safe way to view each observation as "independent" and as part of a temporal pattern (Padoan et al., 2020).

Firstly, the distribution of the variables over the studied period was evaluated through PCA, a widely used chemometric

technique (Jollife and Cadima, 2016). PCA allows rotating the space spanned by the original variables to a new space spanned by the principal components (PCs), in which the first (generally two or three) PCs report the majority of the details found in the original data. As a result, analyzing the behavior of samples (in the scores plot) and variables is made easier by visualizing the two- or three-dimensional plot of PC1 vs. PC2 (and/or PC3) (in the loading plot).

Then, PMF modeling was employed for the allocation of sources and the inner characteristics of ambient air pollutants ($PM_{2.5}$, PM_{10} , CO, O₃, NO₂, and SO₂) on a monthly basis using EPA PMF (version 5.0). PMF also rotates space spanned by the original variables but the calculated factors are not orthogonal to each other like PCA. This way, a better or worse solution can be obtained by PMF modeling by rotating the variables. Standard deviations associated with each measurement are also required to choose the most suitable number of factors and to evaluate the rotational stability of the obtained solution in the PMF analysis (Paatero and Tapper, 1994). PMF allows for the visualization of diverse sources of air contaminants as well as their contributions to specific pollutant concentrations. Several previous research and related sources enumerated in those publications had detailed explanations of PMF algorithms

and their use (EPA PMF 5.0 guide; Sharma et al., 2016a; Cesari et al., 2018; Jain et al., 2020). Therefore, only the compulsory information to elucidate PMF modeling has been described here.

The PMF decomposes a matrix composed of the factors (p), source profiles (f), and the contribution (g) of each source to individual samples based on the following equation (Liu et al., 2017):

$$X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{1}$$

where ij signifies species residuals, i is the number of concentrations, j is the number of chemical components, and X denotes i by j matrix.

Monthly average concentrations of the ambient air pollutants were utilized in the PMF modeling. To attain acceptable results reducing the uncertainties of online datasets, the following precautions were employed: (1) purging of the outliers (far-off values compared to the average value); (2) apportionment of the sources from monthly datasets (using daily mean concentrations obtained from hourly values); and (3) employment of multiple receptor model (Belis et al., 2015; Cesari et al., 2016). Photochemical oxidation of carbon monoxide (CO) and volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x = NO + NO₂) and sunlight is the primary source of ozone (Lu et al., 2018). Thus, including O₃ concentrations could bias the source apportionment of the receptor models by using PMF. Therefore, O₃ was excluded from the input dataset in the modeling.

Standard deviations for each month were included to account for uncertainties of each data point and ease the model to estimate results with a significant confidence level for each pollutant. PMF has the establishment of considering uncertainties accompanying each chemical component of the data (EPA PMF 5.0 guide).

$$Uncertainty = \sqrt{(Error fraction \times Concentration)^2 + (0.5 \times MDL)^2}$$
(2)

where Error fraction is defined as a measure of the standard deviation of particular pollutants divided by the square root of the total number of concentrations. The signal-to-noise (S/N) ratio is utilized to categorize quality of data and the S/N ratios found in this study fall within the acceptable range of those presented in many PMF studies (Sharma et al., 2016a; Cesari et al., 2018; Amato et al., 2020; Jain et al., 2020). The data used in the modeling were specifically divided into three groups based on S/N ratios, with variables with S/N ratios of >2 being labeled as "strong variables," variables with S/N ratios of 0.2-2 being labeled as "weak variables," and the model employing three-fold uncertainty with them. Variables that had S/N ratios <0.2 were disregarded from further analysis and denoted as "bad variables" (Amato et al., 2020). The S/N ratio of all the variables of the air pollutants has been reported in **Supplementary Table 1**.

PMF modeling was employed to run in robust mode, and the Q robust and Q true values were found to be in good agreement with the base run for a four-factor solution. **Supplementary Table 2** displays the Q robust and Q true values for the base runs. The little discrepancies between the two Qvalues indicate that the models were able to achieve a satisfactory fit for the data and outliers. Furthermore, 100% convergence rate for all analytical runs was obtained in the model runs, which also affirms the steadiness of the model and the ability to fit all the variables suitably (Navebare et al., 2018). Extra modeling uncertainty is an additional important parameter that pronounces the fitness of the model. No (0%) error constant (extra modeling uncertainty) was found for all the air pollutants. The results obtained for the air pollutants by PMF modeling had a significant agreement between measured and modeled concentrations specified by R^2 that further confirmed that the concentrations were well-reconstructed (Cusack et al., 2013). R^2 values [from the one-way analysis of variance (ANOVA) F-test] for all the air pollutants were detected to be higher than 0.98, and p-values [from the two-sample Kolmogorov-Smirnov (KS) test] were close to 0 (Supplementary Table 2). The performance factor profiles' uncertainties were also assessed using error estimation methods, namely, the Bootstrap (BS) and Displacement (DISP) methods. Details about error estimation methods used in EPA PMF software, 5.0, have been described by Brown et al. (2015). A strong mapping of the PMF species was observed for all datasets in the BS study, with unmapped cases accounting for <5% of the total. Furthermore, the findings are consistent since no factor profile swap was observed in DISP for any of the datasets (Manousakas et al., 2017).

Human Health Risk Assessment

Adverse effects due to exposure to toxic agents can be inclusively estimated through health risk assessment (USEPA, 1989). This procedure predicts effects on human health caused by a particular pollutant utilizing existing exposure data (WHO, 1996). Exposure to PM_{2.5}, PM₁₀, CO, O₃, NO₂, and SO₂ and their adverse effects has been studied in this study through the US EPA human health risk assessment framework. This method has been employed in several previous studies to assess non-carcinogenic risk due to the criteria pollutants (Megido et al., 2017; Piersanti et al., 2018; Embiale et al., 2020; Mundackal and Ngole-jeme, 2020; Edlund et al., 2021; Morakinyo et al., 2021). The four steps involved in the HHRA are depicted below:

Hazard Identification

Firstly, PM_{2.5}, PM₁₀, CO, O₃, NO₂, and SO₂ have been identified as criteria air pollutants by the WHO as well as USEPA (World Health Organization, 2005; USEPA, 2016).

Dose-Response Assessment

Secondly, the amount of pollutants absorbed by the body was calculated as a function of concentration and exposure time. This research did not have a dose–response analysis. Rather, WHO and the DoE environmental quality guidelines for these air contaminants were used as a benchmark in this study (depicted in **Table 3**).

Exposure Assessment

Then, identification of the exposed population and the magnitude of hazard and duration of exposure are estimated

TABLE 3 Comparison of ambient guideline values of the criteria pollutants in
Bangladesh.

Criteria pollutants	Averaging time	Guideline values				
		Bangladesh ^a	USEPA ^b	WHO ^c		
PM _{2.5} (μg/m ³)	24 h	65	35	25		
	Annual	15	15	10		
PM ₁₀ (µg/m ³)	24 h	150	150	50		
	Annual	50	-	15		
CO (ppm)	1 h	35	35	26.25		
	8h	9	9	9		
O ₃ (ppm)	1 h	0.12	0.12	-		
	8h	0.08	0.08	0.051		
NO ₂ (ppm)	Annual	0.053	0.053	-		
SO ₂ (ppm)	24 h	0.014	0.14	0.0077		
	Annual	0.03	0.029	-		

^acase.doe.gov.bd.

^bwww.epa.gov/criteria-air-pollutants.

^cwww.who.int/news-room/fact-sheets/detail/ambient-(outdoor)-air-quality-and-health.

through exposure assessment. Inhalation is considered the major route of exposure to pollutants. Chronic (annual) and acute (1h) exposure assessments were determined for three different age groups, namely, infants (birth to a year), children (6–12 years), and adults (19–75 years).

The acute exposure assessment for the non-carcinogenic pollutants ($PM_{2.5}$, PM_{10} , CO, O₃, NO₂, and SO₂) was based on the rate equation as follows:

$$AHD = \frac{C \times IR}{BW}$$
(3)

where AHD stands for average hourly dosage for inhalation (μ g/kg/h), C stands for chemical concentration (μ g/m³), IR stands for inhalation rate (m³/hour), and BW stands for bodyweight (kg) (WHO, 1999).

The rate equation for the chronic exposure assessment for the non-carcinogenic pollutants ($PM_{2.5}$, PM_{10} , CO, O₃, NO₂, and SO₂) was:

$$ADD = \frac{C \times IR \times ED}{BW \times AT} \tag{4}$$

where ADD represents the average daily dosage of the chemical of interest ($\mu g/kg/day$), C represents the volume of the chemical in ambient air ($\mu g/m^3$), IR represents the inhalation rate (m^3/day), ED represents the exposure period (days), BW represents the exposed group's body weight (kg), and AT represents the averaging time (days) (WHO, 1999). ED is defined by the following equation:

$$ED = ET \times EF \times DE \tag{5}$$

where ET denotes exposure time (h/day), EF denotes exposure frequency (days/year), and DE denotes exposure period (year). **Table 4** shows the estimated values of each variable used in this assessment for each population group in terms of acute and

chronic exposure periods (Morakinyo et al., 2017, 2021; Embiale et al., 2020).

Risk Characterization

Finally, using the hazard quotient (HQ), the potential noncarcinogenic consequences of exposure to a known pollutant are quantified *via* the "Risk characterization" process (Morakinyo et al., 2017). HQ expresses the likelihood of a negative health outcome among stable and/or sensitive people. The adverse health outcome occurring among different individuals of different age groups is reflected by this process. Thus, noncancer risks associated with the air pollutants were estimated through HQ. Acute and chronic non-cancer risks were calculated according to the following equations:

$$HQ = \frac{ADD}{RfC} \ \left(chronic \ exposure \right) \tag{6}$$

$$HQ = \frac{AHD}{RfC} (acute \ exposure) \tag{7}$$

where RfC is an approximation containing uncertainty of considerable scale of an incessant inhalation exposure to the human population (including sensitive subgroups) that possesses a substantial risk of lethal effects during a lifetime (www.epa.gov). The chemical identifications, RfC values, and affecting organs are presented in **Supplementary Table 2**.

An HQ value of 1.0 indicates no risk to human health, while an HQ value of 1.0 indicates a marginal risk, indicating that the considered contaminant is not a potential health risk, to even a sensitive person. However, an HQ > 1.0 signifies risks to some extent upon exposure to different individuals, adults, and/or children (USEPA, 1989).

RESULTS AND DISCUSSION

Ambient PM Trends in Dhaka

Long-term yearly as well as monthly trends of PM_{2.5} and PM₁₀ have been depicted in Figure 3. Supplementary Table 4 shows the statistics of PM_{2.5} and PM₁₀ during the studied period. Both PM_{2.5} and PM₁₀ showed normal distribution for the studied period with mean yearly concentrations of 88.69 \pm 9.76 and $124.57 \pm 12.75 \ \mu g/m^3$, respectively. The yearly values of PM_{2.5} and PM₁₀ exceeded the national air quality standards respectively by nearly 6.0 and 2.5 times while exceeding WHO standards by 9.0 and 6.0 times. The linear regression analysis has been performed to determine the trends and statistical significances of PM_{2.5} and PM₁₀. Increasing trends have been observed for PM_{2.5} and PM_{10} over the years with slopes of 1.83 \pm 0.15 and 2.35 \pm $0.24 \,\mu g/m^3$ /year, respectively. One-way ANOVA showed that the increasing trends were statistically significant (p < 0.05) for both the PMs (Figure 3). Rana et al. (2016) reported similar findings in Dhaka that showed that PM2.5 concentrations exceeded the WHO guideline value by 8-13 times. Begum and Hopke (2018) also showed that long-term trends for PM2.5 and PM10 exceed the USEPA standards in Dhaka.

Monthly variations were identical for the PMs with the highest values observed in winter (November–February) and the lowest

TABLE 4 | The variables used in the calculation of exposure rate and risk assessment factors for different age groups.

Variable	Description	n Value						
		Infant (Bi	Infant (Birth to 1 Year)		Child (6-12 years)		Adult (19–75 years)	
		Acute	Chronic	Acute	Chronic	Acute	Chronic	
С	Contaminant concentration in ambient air							μg/m ³
IR	Mean inhalation rate	0.30	6.80	1.20	13.50	1.20	13.30	m³/h
BW	Mean body weight	11.30		45.30		71.80		kg
ET	Exposure time	1.00	14.60 [(350/24) × 1]	1.00	1050.00 [(4200/24) × 6]	1.00	1312.50 [(10,500/24) × 3]	h
EF	Exposure frequency	350.00		350.00		350.00		days
DE	Exposure duration	1.00		12.00		30.00		years
AT	Averaging time	365.00 (1 × 365)		4380.00 (12 × 365)		10,950.00 (30 × 365)		days

in monsoon (June–August). Previous studies also supported such seasonality of PMs with the highest values being associated with the dry season compared to the wet season (Rana et al., 2016; Rahman et al., 2020).

Gaseous Pollutants' Trends in Dhaka

Trace gases CO, O₃, NO₂, and SO₂ showed yearly concentrations of 0.69 \pm 0.06 ppm, 51.42 \pm 1.82 ppb, 14.87 \pm 2.45 ppb, and 8.76 \pm 2.07 ppb, respectively for 2003–2019. Normal distribution was observed for all the trace gases over the studied period (**Supplementary Figure 1**). NO₂ and SO₂ were below the annual national guideline values set by DoE. Yearly trends of CO, O₃, NO₂, and SO₂ showed slopes of 0.01 \pm 0.002 ppm/year, 0.13 \pm 0.09 ppb/year, 0.47 \pm 0.03 ppb/year, and 0.40 \pm 0.02 ppb/year for the studied period. One-way ANOVA showed that the trends of the trace gases were statistically significant (p < 0.05) except for O₃ (**Figure 4**). Rahman M. M. et al. (2019) showed similar significant positive trends for SO₂ and CO in Dhaka, Bangladesh for 2013–2017.

Monthly observations showed that CO, NO_2 , and SO_2 followed an almost similar pattern over the year with the highest values in winter (November–February) and the lowest in monsoon (June–August). On the contrary, O_3 showed the maximum values in March–May and the minimum in July–September. The seasonal trends of ozone in Dhaka differed from those of other contaminants, which all exhibited distinct seasonal variation with a peak in the winter and a trough in the monsoon (Rahman M. M. et al., 2019).

Meteorological Parameters' Trends in Dhaka

The air quality of an urban area is heavily influenced by meteorological factors (Rahman M. S. et al., 2019; Afrin et al., 2021). Long-term trends of the meteorological parameters have been depicted in **Figure 5**. Surface temperature, pressure, and relative humidity showed yearly values of $25.99 \pm 0.750^{\circ}$ C, 1006.46 \pm 0.54 hPa, and 71.30 \pm 1.36%, respectively, over

the studied period. Monthly variation of temperature showed the highest values in April–June and lowest in December– January. Surface pressure showed opposite monthly variation with the highest values observed in June–July and lowest in December–January. Relative humidity, on the other hand, showed the highest values in June–September and lowest in February–March. Linear regression of the parameters showed slopes of $0.12 \pm 0.02^{\circ}$ C/year, 0.03 ± 0.02 hPa/year, and $0.11 \pm$ 0.07%/year, respectively, for temperature, pressure, and RH. The one-way ANOVA study revealed that the trends were statistically insignificant except for temperature.

Correlation Among the Pollutants and Meteorological Parameters

Pearson correlation analyses were employed to quantify relationships among the ambient air pollutants and meteorological parameters as the variables did not violate the normality test (**Table 5**; **Supplementary Figure 1**). $PM_{2.5}$ and PM_{10} showed significant positive values with PMs, NO₂, and SO₂. Furthermore, NO₂ and SO₂ also showed significant positive values with each other, PMs, and CO. The significant positive correlations among the air pollutants suggested similar production pathways for them.

Previous studies also investigated associations among the air pollutants and meteorological parameters. Ozone had nonsignificant associations with the other pollutants, according to Rahman M. M. et al. (2019). Although not significant, O₃ has a positive association with PMs, CO, NO₂ ($R^2 > 0.37$), suggesting that the other pollutants could play important roles in the development of O₃. Besides, O₃ showed a negative correlation with RH but a positive correlation with temperature.

In the case of the meteorological parameters, temperature showed significantly positive correlations with all the pollutants but not in the case of the other meteorological variables. Surface pressure did not show any significant association with any other variables other than RH. Lastly, RH did not exhibit any significant connection with the variables. However, according to Afrin et al.



regions represent the standard deviations).

(2021), air temperature, wind speed, and wind direction could account for more than 90% of $PM_{2.5}$ variability.

Source Apportionment

Principal Component Analysis

PCA was employed on the monthly datasets of the air pollutants. The datasets were auto-scaled before the analysis. This indicates that the column mean has been subtracted from each digit of the dataset, and the result has been separated by the column standard deviation. **Figure 6** shows the bi-plots obtained by the PCA carried out on the datasets. It reports both scores and loadings in the same PC1 vs. PC2 graph. PC1 and PC2 loadings account for 98.88% of the total variance of the pollutants. PC1 carries 45.20, 45.20, 45.02, 44.09, and 44.09% of explained variance for PM_{2.5}, PM₁₀, CO, NO₂, and SO₂ and PC2 carries 55.32 and 55.25% of explained variance for NO₂ and SO₂ (**Figure 6**). The

two PMs, CO, NO₂, and SO₂ have the same characteristics as they are distributed evenly along the *x*-axis, which accounts for almost ~45% of the variance. Thereby, PC1 can be inferred to be vehicular emissions according to the component variance test, whereas NO₂ and SO₂ showed similar discrimination along the *y*-axis, contributing ~55% of these variables. Thus, PC2 can be attributed to the industrial emissions enriched in NO₂ and SO₂. Furthermore, the variables are almost evenly distributed in the positive and negative regions of PC1 and PC2. It indicates that the chemical composition of the studied compounds has similar concentrations in the case of these two variables.

PMF Modeling

The factor profiles of all the sources of the air pollutants ($PM_{2.5}$, PM_{10} , CO, NO_2 , and SO_2) are depicted in **Supplementary Figure 2**. Four factors were attributed to be



FIGURE 4 | Yearly and monthly trends (A) of the gases (SO₂, NO₂, O₃, and CO) and the long-term evolution of SO₂, NO₂, (B) O₃, and CO (C) for 2003–2019 (the shaded regions represent the standard deviations).



humidity (C) for 2003–2019 (the shaded regions represent the standard deviations).

Pearson correlation	PM _{2.5}	PM ₁₀	со	O ₃	NO ₂	SO ₂	т	Р	RH
PM _{2.5}	1.00	0.96**	0.93**	0.42	0.96**	0.96**	0.69**	0.33	0.34
PM ₁₀	0.96**	1.00	0.96**	0.43	0.94**	0.96**	0.68	0.25	0.22
СО	0.93**	0.96**	1.00	0.37	0.87**	0.92**	0.61*	0.22	0.18
O ₃	0.42	0.43	0.37	1.00	0.42	0.41	0.41	0.05	-0.31
NO ₂	0.96**	0.94**	0.87**	0.42	1.00	0.95**	0.66**	0.37	0.40
SO ₂	0.96**	0.96**	0.92**	0.41	0.95**	1.00	0.78**	0.25	0.29
Т	0.69**	0.68**	0.61*	0.41	0.66**	0.78**	1.00	0.22	0.22
Р	0.33	0.25	0.22	0.05	0.37	0.25	0.22	1.00	0.50*
RH	0.34	0.22	0.18	-0.31	0.40	0.29	0.22	0.50*	1.00
RH									

TABLE 5 | Pearson correlation coefficients among the air pollutants (PM_{2.5}, PM₁₀, CO, NO₂, and SO₂) and the meteorological parameters (surface temperature, pressure, and relative humidity).

*Significant at the 95% confidence level.

**Significant at the 99% confidence level



the estimated sources contributing to the aforementioned ambient air pollutants. The percent contributions of the factors influencing the air pollutants have been depicted in **Figure 7**.

Factor 1: The first factor is characterized by the highest contribution (74.7%) to NO₂ followed by the signature of 36.5, 35.7, 20.5, and 19.7%, respectively, for PM_{2.5}, PM₁₀, CO, and SO₂ (**Figure 7**). Thus, factor 1 is inferred to be vehicular emissions as these pollutants were significantly correlated, suggesting similar sources. Despite the fact that many buses in Dhaka have been converted to CNG engines, heavy-duty trucks that are limited continue to run on diesel between the hours of 10 p.m. and 6 a.m. However, the lower contribution to SO₂ suggests that the conversion of most light-duty vehicles and buses to CNG has resulted in significant reductions in light-duty vehicle and bus emissions (Begum and Hopke, 2018).

Factor 2: PM loadings appeared to be highest (42.9 and 42.7%, respectively, for $PM_{2.5}$ and PM_{10}) for factor 2 with contributions

of 28.5 and 22.5% for CO and NO₂, suggesting that this factor can be inferred to be road/soil dust or secondary sources. Elemental characterization is needed to correctly attribute this factor to the ambient pollutants. Spearman correlation revealed that PMs have significant correlations with CO, NO₂, and SO₂ that further affirms traces of the same factor in the case of these pollutants. Due to both anthropogenic and natural sources, dust has been identified as a significant contributor to measure PM_{2.5} in Dhaka, accounting for 11% of total PM_{2.5} (Weagle et al., 2018).

Factor 3: This factor is characterized by the highest contribution (50.5%) to CO with significant contributions to SO₂. PMs are also influenced by this factor with loadings of 14.2 and 15.1%, respectively, for PM_{2.5} and PM₁₀. This factor can be inferred to biomass or fossil fuel burning as suggested by the high loading of CO. Ommi et al. (2017) reported the transboundary influence of biomass burning from the IGP to be an important factor in Dhaka, mainly arising from the preparations of the lands through field burning. Furthermore, Salam et al. (2008) reported that the higher concentration of SO₂ in the city center is possibly due to the high content of sulfur in fossil fuel.

Factor 4: Factor 4 can be inferred as industrial emissions as it contributes mostly (39.2%) to SO₂ with traces in the case of CO (0.5%) and NO₂ (2.0%). Both PM_{2.5} and PM₁₀ are associated with about 6.5% for factor 2. The high contribution to SO₂ from this factor signifies the effect of industrialization centered in Dhaka, which has increased exponentially in the last decade or so. The commercial areas of Dhaka had the highest SO₂ concentration (76.8 μ g/m³) (Salam et al., 2008).

However, since sources of atmospheric pollutants are extremely complicated and diverse, particularly for unorganized emission sources, ambient pollutants cannot be apportioned perfectly through receptor modeling, and some factors remain unknown (Liu et al., 2016, 2017). Furthermore, the dearth of element analysis for metals limits the capability of receptor models to categorize all the sources present regionally. Moreover, the transboundary effects are also significant for ambient regional pollutants in Bangladesh that might affect the ambient air quality as reported by many previous studies (Rana et al., 2016; Rahman et al., 2020).



Human Health Risk Assessment

The HQ was calculated for three age groups (infant, child, and adult) in acute and intermediate level exposures, which have been reported in Figure 8. At the acute exposure level, no pollutants showed an adverse effect, that is, HQ values were below 1, for any of the age groups. On the other hand, the HQ values posed significant health risk (HQ > 1) at the chronic exposure level for infants and children while no antagonistic health effects risk (HQ < 1) were observed for the pollutants in the case of the adults. Among the pollutants, PM_{2.5} (1.81 \times 10^3 \pm 2.00 \times $10^2)$ and O_3 (1.03 × 10³ ± 3.65 × 10¹) were observed to be the most harmful pollutants affecting the studied age groups, especially the children. In terms of the studied age groups, children were observed to be affected far more than infants and adults at the chronic exposure level. These results suggest that the air pollutants affect all age groups significantly when considered at the chronic level with children being the worst sufferers while negligible effects are observed at the acute level. Owing to a variety of factors, including their comparatively higher level of air inhalation (a resting infant's air consumption per weight unit is double that of an adult), their immune system and lungs not being fully grown, children are the population most impaired by indoor air contamination (Lina Thabethe et al., 2014). Morakinyo et al. also demonstrated a similar adverse impact on children when compared to other age groups. These results essentially signify that low breathing heights could be the main factor along with other indoor and outdoor air pollution sources behind this phenomenon in children who are particularly susceptible to ground-level pollution. In a study conducted at a local school, Sharma and Kumar (2020) found that in-pram babies are exposed to up to 44% higher fine particle concentrations than adults. **Table 6** lists some more previous studies comparing health risks associated with ambient air pollutants.

DISCUSSION AND FUTURE IMPLICATIONS

The air pollutant ($PM_{2.5}$, PM_{10} , CO, NO_2 , and SO_2) concentrations except O_3 showed significantly increasing trends in this study. However, an analysis of Dhaka's air quality over two decades (1996–2015) found that the city's air quality has remained constant over the last decade, despite increased economic development and the number of sources such as passenger cars and brick kilns (Begum and Hopke, 2018). On the contrary, a recent analysis found that $PM_{2.5}$ concentrations in Dhaka decreased slightly (statistically non-significant) from 2013 to 2017, but fine PM concentrations remained elevated and



Study area	Studied variables	Findings	References
Gijón, Spain	PM ₁₀ samples	Cancer and non-cancer risk values were within tolerable limits for both adults and children while the health risk was predicted to be higher for children	Megido et al., 2017
Cordoba city, Argentina	PM samples	HI < 1 for all age groups and land use areas	Mateos et al., 2018
Beihai and Shanghai, China	Heavy metals in street dust	Non-carcinogenic risks due to Cr for children in both the sites were relatively higher than other metals (HI < 1)	Chen et al., 2019
Dhaka, Bangladesh	Heavy metals in street dust	Children may have a slight non-cancer health risk as a result of exposure to street dust (HI $>$ 1).	Rahman M. S. et al., 2019
Dhaka, Bangladesh	Heavy metals in road dust and roadside soil from different school buildings	Children are more susceptible to non-cancer risks than adults and ingestion was identified as the dominant pathway (HI < 1).	Rahman et al., 2021
This study	$PM_{2.5},PM_{10},CO,O_3,NO_2,andSO_2$	HQ < 1 at acute levels but $HQ > 1$ for infants and children at chronic level	

presumably continue to affect human health (Rahman M. M. et al., 2019). Therefore, there remains a need for further study into long-term seasonal cycles, as well as their assessment concerning actions taken by relevant bodies.

Among the meteorological parameters, the temperature seemed to significantly correlate with the air pollutants ($PM_{2.5}$, PM_{10} , CO, NO₂, and SO₂) except O₃. However, according to Afrin et al. (2021), meteorological factors (temperature, relative humidity, etc.) account for about 57% and 35% of the variations

in PM_{2.5} and PM_{2.5-10} concentrations, respectively, implying that the finer PM fraction is affected more by meteorology than the coarser fraction. Thus, more quantitative approaches to how climate change affects urban air quality may provide further insights into contaminant reduction policies.

This study estimates four probable main sources contributing to the emission of the six air pollutants in Dhaka, Bangladesh, which are vehicular emissions, road/soil dust, biomass burning, and industrial emissions. In a study on NO_x pollution from

vehicular emissions in Dhaka, Iqbal et al. (2019) found that, while a critical urban health ecosystem already exists, the current pattern of vehicular expansion, combined with current vehicle technology and on-road traffic management systems, may soon lead to an unbecoming situation. As a result of these findings, the vehicular management system in Dhaka should be strengthened, and further research into PMs, CO, and NO_x emissions caused by them should be conducted. Furthermore, according to Rahman M. S. et al. (2019), ingestion of dust particles increases the risk of heavy metals (Cr, Cd) in children and adults in Dhaka City, where children were facing a possible health risk. Dhaka is witnessing several infrastructure schemes, such as the Dhaka Metrorail Project and the Dhaka Elevated Expressway Project, which have raised the risk of dust contamination by a factor of many. So, heavy metal contamination caused by dust particles should be investigated more thoroughly in Dhaka, as it poses a threat to children in particular. Rahman et al. (2020) suggested that air pollution in Dhaka is influenced by both local and transboundary sources, but biomass-related PM2.5 was found to be the most prevalent during cycles of crop-burning in the IGP. Thus, biomass burning is a major contributor to Dhaka's poor air quality, necessitating further study into the source's impact and mitigation. Finally, the rising pattern of SO₂ in this study and previous studies (Rahman M. M. et al., 2019) indicates that industrial operations in Dhaka, especially brick kiln operations and high sulfur diesel usages, are still contributing significantly to detrimental air quality. More studies encompassing the brick kiln operations and other industrial usage of high sulfur materials in Dhaka are also of paramount importance. Begum et al. (2011) described vehicular emissions and emissions from brick kilns as the two major sources of air pollution in Dhaka and stated that the government of Bangladesh is exploring various measures to minimize emissions from those sources through the adoption of regional policies. However, the pollutant patterns in this study indicate that effective implementation of such measures is still scarce in Dhaka, Bangladesh.

CONCLUSION

In this study, monthly ambient concentrations of the air pollutants (PM_{2.5}, PM₁₀, CO, O₃, NO₂, and SO₂) and meteorological data (temperature, surface pressure, and relative humidity) were utilized to explore long-term variations of these parameters and their association with each other. Long-term trends of the ambient pollutants were observed to be increasing significantly in the one-way ANOVA test except for surface ozone while the meteorological parameters showed no significant trends over the studied period. Pearson correlation studies revealed that the ambient pollutants were significantly (CI > 95%) correlated with each other, suggesting probably the same sources. The PCA bi-plot revealed that the PMs and CO demonstrated similar variance while NO₂ and SO₂

followed an analogous pattern. However, four factors emerged as estimated sources of the pollutants in PMF receptor modeling, which were vehicular emissions, road/soil dust, biomass burning, and industrial emissions. PMs were dominated (\sim 42%) by the road/soil dust along with emissions from vehicles. Biomass burning played a major role in CO (50.5%) and SO₂ (41.1%) production while industrial emissions were another prominent factor (39.2%) in the case of SO₂. On the contrary, NO₂ was identified to be mainly emitted from vehicular emissions (74.7%).

Health risk assessment of the pollutants for three age groups (infant, child, and adult) in acute level exposures indicated no adverse effect (HQ < 1) for any of the age groups. On the other hand, the HQ values posed significant health risk (HQ > 1) at the chronic exposure level for infants and children while no antagonistic health effects risk (HQ < 1) were observed for the pollutants in the case of the adults.

This study reveals that the implementation of CNG wheelers in 2003 might have reduced Pb emissions in megacity Dhaka, but vehicular emissions along with road dust, biomass burning, and industrial emissions remain the most prominent sources that have a significant hazard risk on children and infants particularly. However, since most studies have focused on Dhaka, there is still room to assess long-term possible health risks and the effect of climate variables on air quality at the divisional and district levels in Bangladesh.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct and intellectual contribution to the work, and approved it for publication.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/frsc.2021. 681759/full#supplementary-material

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Air Pollution, Climate Change, and Human Health in Indian Cities: A Brief Review

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Climate change and air pollution have been a matter of serious concern all over the world in the last few decades. The present review has been carried out in this concern over the Indian cities with significant impacts of both the climate change and air pollution on human health. The expanding urban areas with extreme climate events (high rainfall, extreme temperature, floods, and droughts) are posing human health risks. The intensified heat waves as a result of climate change have led to the elevation in temperature levels causing thermal discomfort and several health issues to urban residents. The study also covers the increasing air pollution levels above the prescribed standards for most of the Indian megacities. The aerosols and PM concentrations have been explored and hazardous health impacts of particles that are inhaled by humans and enter the respiratory system have also been discussed. The air quality during COVID-2019 lockdown in Indian cities with its health impacts has also been reviewed. Finally, the correlation between climate change, air pollution, and urbanizations has been presented as air pollutants (such as aerosols) affect the climate of Earth both directly (by absorption and scattering) and indirectly (by altering the cloud properties and radiation transfer processes). So, the present review will serve as a baseline data for policy makers in analyzing vulnerable regions and implementing mitigation plans for tackling air pollution. The adaptation and mitigation measures can be taken based on the review in Indian cities to reciprocate human health impacts by regular air pollution monitoring and addressing climate change as well.

Keywords: air pollution, climate change, aerosols, urban, India, health

INTRODUCTION

Air pollution and climate change are major threats to rapidly growing cities in present times. The developing nations like India, which are switching from predominantly rural country to increasingly urban, have to face critical challenges in terms of climate action and sustainable development (Van Duijne, 2017; Singh C. et al., 2021). India is projected to have 53% of the national population as urban population by addition of 416 million urban dwellers by the year 2050 (UNDESA, 2018).

The change in land use land cover patterns in urban areas due to ongoing urbanization affects regional climate by altering the surface and boundary layer atmospheric properties (Shepherd, 2005; Ren et al., 2008; Yang et al., 2012). Further, the urbanization by changing land use land cover affects climate via increased anthropogenic emissions, extreme precipitation (that may cause

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Kaur R and Pandey P (2021) Air Pollution, Climate Change, and Human Health in Indian Cities: A Brief Review. Front. Sustain. Cities 3:705131. doi: 10.3389/frsc.2021.705131 urban flooding), higher temperatures, and frequent heat waves with heat related human health impacts (Chestnut et al., 1998; Ramanathan et al., 2001; Shastri et al., 2015). The regional climate changes are reflected by different meteorological conditions such as changes in temperature and precipitation. The anthropogenic emissions such as greenhouse gas (GHG) emissions trigger these local climate changes.

In addition to the impact of urbanization on climate, the increasing urban population and vehicular traffic increases the pollutant emissions and aerosol load in the atmosphere. The increasing urbanization alongwith growing population and industrialization has been stated as one of the key reasons for high aerosol loading in the Indian sub-continent (Kaskaoutis et al., 2011; Ramachandran et al., 2012; Krishna Moorthy et al., 2013). Thus, the climate change and air pollution remain one of the biggest threats to human health and well-being in cities and are interlinked with each other as discussed later in this study.

According to a report by World Health Organization (WHO) more than seven million people across the world lose their lives due to diseases linked with PM_{2.5} pollution (WHO, 2015). India, being a rapidly developing country with increasing population is suffering from severe air pollution; as among the world's 10 most polluted cities, nine of them lie in India [WHO Global Urban Ambient Air Pollution Database (Update 2016), 2016]. The increasing air pollution in most of the Indian megacities over last few decades and its consequential human health impacts (such as asthma and cardio-respiratory illness) have drawn prominent attention in recent years (Sarath and Ramani, 2014; Gautam et al., 2020; Shaw and Gorai, 2020).

CLIMATE CHANGE

The global change in climate has been reported by various workers in the last few decades. The natural process of climate change because of volcanic eruptions, continental drift, and astronomical cycles is now accelerated by human activities (IPCC, 2007). The emission of greenhouse gases (GHGs) is one of the major factors in altering climate by changing atmospheric concentration of certain gases. Further, the role of water vapors in altering the climate is also being well looked into by scientists (Jacob, 2001; Forster and Collins, 2004). Not only this, scientists are also looking into the role of black carbon in climate change due to their ability to strongly absorb incoming solar radiations (Jacobson, 2001; Ramanathan and Carmichae, 2008; Surendran et al., 2013). Menon et al. (2002) used a global climate model to investigate the role of aerosols in altered climate in India and China and reported that precipitation and temperature changes in the model could be correlated to large load of absorbing black carbon in the aerosols. Also, due to heating of air by black carbon aerosols, atmospheric stability is altered leading to changes in hydrologic cycle and large-scale circulations.

Climate change is known to alter the temperature, precipitation pattern and solar insolation over the planet. According to IPCC (2007) report, about 0.65°C increase has been observed in global average surface temperature over last 50 years and is projected to increase by 1.1–6.4°C. The rise

in sea level has been observed with ongoing warming trend. Annual sea level rise between 2.5 and 3 mm along the coastline of Mumbai has been reported (Pramanik, 2017). Also, according to a study by NASA, this region has possessed increase in average temperatures by 2.4°C for the period from 1881 to 2015 (NASA, 2015). Further, an increase in frequency of extreme rainfall was analyzed that can cause flooding. This can also be seen in Mumbai, one of the Indian mega-city and home to the largest population threatened by coastal flooding (Intergovernmental Panel on Climate Change IPCC-SREX, 2012). Mumbai has been recognized as one of the world's most vulnerable cities to climate change according to UN-HABITAT, 2010 (Mehta et al., 2019). The changes in rainfall in the past century (1901-2019) were observed over India by Kuttippurath et al. (2021). In the study of 119 years of rainfall measurements, a significant change in the rainfall pattern has been confirmed after 1973 with the declining trend of about 0.42 ± 0.024 mm dec⁻¹. The study shows that in recent decades, the wettest place of the world has shifted from Cherrapunji to Mawsynram.

Besides, the increasing temperature due to climate change can trigger melting of glaciers. A study conducted by Kumar et al. (2021) for monitoring glacier changes in Nanda Devi region of Central Himalaya, India, for three decades, shows the loss of about 26 km² (10%) of the glaciated area between 1980 and 2017. Additionally, the climate change causing extreme weather events causes increase in frequency and intensity of floods, storms, torrential rains, and droughts etc. that take thousands of lives and affect millions of people (Haines et al., 2006; Majra and Gur, 2009). The projected climate change estimates from 2036 to 2060 for 57 Indian cities show that 33 cities are likely to experience rise in extreme rainfall and exacerbated flood risk. The remaining 24 cities will observe precipitation declines, reflecting higher drought risk (Ali et al., 2014; Singh C. et al., 2021).

The land use land cover being an important factor in climate change has been focused in many studies over Indian cities such as Nath et al. (2021) that shows rapid expansion of built-up areas in Guwahati with an overall increase of 103% in area over the last 30 years (1990–2020). The expansion in urban areas causes decline in vegetated areas, cropland, and fallow land thereby contributing to climate change. Paul et al. (2021) also showed expansion in urban areas at annual rates of 38.6% with decline in agricultural area at rate of 2.1% for peripheral Delhi during the 1973–2017. So, climate change is one of the emerging threats to human health in Indian cities. With the increase in climate variability, there is an associated increase in health issues (Bush et al., 2011). Cities, due to UHI occurrence, are supposed to have higher effect of climate extremes such as precipitation extremes and heat waves than rural regions (Shepherd, 2005; Shastri et al., 2017; Chauhan and Singh, 2020).

Human Health Impacts Associated With Climate Change

The adverse impacts of climate change on human health have been documented in several studies and these effects are expected to increase with future climate change (Luber and McGeehin, 2008; Bell et al., 2018; Filippelli et al., 2020). The climate change affects human health by problems induced from notable extreme weather conditions such as increased temperature, precipitation, increased intensity and frequency of heat waves, floods, droughts, strong winds, and landslides (Orimoloye et al., 2019). The change in temperature and precipitation causing severe heat, extreme cold, and unpredictable rain linked with climate change increases health related issues; as these climatic changes further induces water and air-borne infections, vector borne-infections, malnutrition, incidence of diarrhoeal diseases, and heat related morbidity and mortality (Haines et al., 2006; Dutta and Chorsiya, 2013).

Children, elderly people, and urban residents are more vulnerable to these health impacts (Haines et al., 2006; Ebi and Paulson, 2010; Filippelli et al., 2020). Nearly 150,000 deaths and about 5 million illnesses have been reported per year due to climate change (Dutta and Chorsiya, 2013).

The respiratory infections, chronic obstructive pulmonary disease, pneumothorax, asthma, allergies, hyperthermia, and dehydration are some of human health issues associated with climate change either directly or indirectly (D'Amato et al., 2011; Filippelli et al., 2020). About 6% of children in India are prone to respiratory tract infections and 2% of adults in India are also trapped in asthma disease (International Institute for population sciences (IIPS) Macro International, 2007).

Thus, these extreme weather conditions have adverse health impacts that can also result in loss of lives. If we look at the extreme heat related human health effects, it becomes imperative to understand the effects of rising temperature on biota.

The increase in temperature due to climate change is a major cause of heat-related diseases in cities such as skin cancer, heat stroke, heart disease, diarrhea, and increased mortality (Changnon et al., 1996; Hondula and Barnett, 2014; Orimoloye et al., 2019). The heat related human health impacts also include dehydration, heat cramps, heat exhaustion, heat stroke, loss of fluids, heat injuries, eye, and skin diseases (Dutta and Chorsiya, 2013). The increase in urban temperature or the urban heat island (UHI) effect is an important implication of climate change. The UHI effect have been reported in many Indian cities (Ambinakudige, 2011; Kikon et al., 2016; Mathew et al., 2016; Kaur and Pandey, 2020) causing thermal discomfort to urban residents. This effect is linked with certain respiratory problems due to deterioration of air quality by cooling agents (Liu and Zhang, 2011).

Besides, the heat waves along with other frequent weather events are reported as significant evidence of climate change in eastern India (Patil and Deepa, 2007). The heat wave during 1998 and 2015 has taken lives of more than 2000 people each in India (Mukherjee and Mishra, 2018). Approximately 1,625 people lost their lives in Rajasthan, followed by Bihar, Uttar Pradesh, and Odisha during 1978 to 1999 due to heat wave (De, 2000); while the toll increased to 3,442 heat-related deaths during 1999–2003 (Chaudhury et al., 2000; Centers for Disease Control Prevention, 2006). The statistical data documented in a study by Dutta and Chorsiya (2013) states that more than 600 people have died due to heat wave in India in 2013. Besides, about 1,400 deaths were caused by high ambient temperature (50°C) in Andhra Pradesh in 2002. Similarly, in Ahmadabad high ambient temperature (46.8°C) took lives of many urban residents in 2010. Further, the heat waves significantly affected dozens of Indian states such as Rajasthan and Uttarakhand in 2009 (Dutta and Chorsiya, 2013). The climate variability and its relation with mortality due to heat in India were documented by Akhtar (2007), Dholakia et al. (2015) for Ahmedabad, Murari K. K. et al. (2015), and Mazdiyasni et al. (2017). Further, following the increasing frequency of hot days and nights during the period 1951–2016, 4-fold increase has been projected by 2050 and 12fold by 2,100 that will lead to increased heat-related mortality (Mukherjee and Mishra, 2018; Singh C. et al., 2021).

The climate change is known to trigger other extreme events such as drought, floods, tsunamis, and cyclones that are associated with negative human health impacts. Urban drought and floods caused by changing climate due to scarcity or excess of rainfall indirectly affects human health. Drowning, hypothermia, and trauma are some physical effects of floods on human health (Ahern et al., 2005; Du et al., 2010). Severe drought conditions resulting in scarcity of food caused high number of deaths due to starvation in India (Dutta and Chorsiya, 2013). Also, the high rainfall causing floods lead to destruction of crops that in turn causes shortage of food leading to malnutrition and public health issues. The malnutrition is a severe issue in India with about 47% of the children prone to this problem according to World Bank Report on Malnutrition in India (2009). The malnutrition can further cause anemia from which about 70% children, 55% women, and 25% of men population are suffering, in India (Majra and Gur, 2009).

The rising sea level due to climate change may cause flooding that can cause death (Dutta and Chorsiya, 2013). Moreover, the drought and flood conditions also decrease the availability of fresh water. The increase in risk of diarrheal diseases linked with floods has been reported for India (Mondal et al., 2001). The contaminated water can cause transmission of various waterborne infections leading to *E. coli* infection, cholera, typhoid, cryptosporidium, shigella, giardia, and viruses such as hepatitis A (Gabastou et al., 2002; Kovats and Akhtar, 2008; Majra and Gur, 2009). Besides, floods also lead to certain rodent-borne diseases such as including tularemia, leptospirosis, and viral hemorrhagic diseases. Lyme disease, Hantavirus pulmonary syndrome, and tick-borne encephalitis are some other diseases linked with climatic variability for Baltimore and London (Wilson et al., 2001; Majra and Gur, 2009).

Besides, the extreme weather events due to climate change such as cyclones, tsunamis, and floods have taken thousands of lives and affected millions of population in many Indian states such as Assam, Bihar, West Bengal, Odisha, Uttar Pradesh, Himachal Pradesh, Rajasthan, and Gujarat (World Health Organization (WHO), 2005; Majra and Gur, 2009). These events also cause adverse health impacts on surviving population. Some of these extreme weather events reported in the past few years are:

Heat wave in Odisha in 1998 and 2004, Super cyclone in Odisha with wind speed over 300 km/h in October 1999, Heat wave in Andhra Pradesh in 2003, Cold wave in Uttar Pradesh and Uttaranchal in 2004, Tsunami affecting Tamil Nadu, Andhra Pradesh, Kerala, and the Andaman-Nicobar Islands in 2004,

Heaviest rainfall in Indian metropolitan city of Mumbai in 2005, Cyclone in Andhra Pradesh in 2005, Floods in Gujarat and Madhya Pradesh in 2005 and cloudburst and flood in Uttarakhand in June 2013.

These disasters enhances the incidence and spread of diseases by increasing transmission of infectious vectors such as plague, Japanese encephalitis, malaria, dengue fever, chikungunya, and filariasis (Bhattacharya et al., 2006; Devi and Jauhari, 2006; Dhiman et al., 2008; Bush et al., 2011). Additionally, these calamities have badly affected Indian states such as Plague in Surat, malaria in Odisha, West Bengal, Jharkhand, Chattisgarh, Madhya Pradesh, and North East (Kumar et al., 2007). The coastal regions of India are prone to tsunamis and cyclones (Dutta and Chorsiya, 2013).

These disasters also lead to occurrence of water-borne diseases such as amoebiasis, crytosporidiosis, giardiasis, typhoid, cholera, and other infections. According to The World Health Organization (WHO), 900,000 Indians die each year from drinking contaminated water and breathing polluted air (World Health Organization and United Nations Children's Fund (WHO and UNICEF), 2000). Also, Indian Ministry of Health estimated 1.5 million deaths annually between 0- and 5-year-old children. Every year in India around 0.6–0.7 million children under 5 years of age die from diarrhea.

So, the potential health impacts related with climate change can be categorized as:

- a) Direct factors: The factors such as thermal stress, death/injury in floods and storms are direct implication of climate change that affects human health.
- b) Indirect factors: The indirect factors include vectors-borne diseases, water-borne pathogens, water and air quality, and food availability and quality that are indirectly caused by climate change.

AIR POLLUTION

Air pollution is a matter of serious concern in megacities where the pollution levels often exceed the permissible limits due to its associated health risks for city residents (Chattopadhyay et al., 2010; Debone et al., 2020). The metropolitan cities of India are exposed to unhealthy and unhygienic conditions due to air pollution (Dutta et al., 2021). The continuous and alarming increase of urban air pollution is an emerging environmental issue in the Indian megacities for the last few decades (Faheem et al., 2021).

Major outdoor and indoor air pollutants in urban areas can be primary or secondary air pollutants. Primary air pollutants that are emitted directly include particulate matter $\{PM_{2.5}, PM_{10}, suspended particulate matter (SPM), respirable$ $particulate matter (RPM)\}, SOx, NOx, CO, ammonia, and dust$ particles while the secondary air pollutants involve ozone, smog,Peroxyacyl nitrates (PANs) etc.

The developing nations like India with ongoing urbanization are suffering from increased air pollution issues due to the lack of services such as adequate transportation management, suitable roads, and unplanned distribution of industries (Rumana et al., 2014). The congested roads in cities reduce average vehicular speed resulting in higher vehicular emissions adding to air pollution levels (West Bengal Pollution Control Board, 2010). The increasing and unplanned urbanization coupled with industrialization and population growth are posing threat to human health by increasing air pollution levels leading to number of health issues (Dutta et al., 2021). Additionally, the complex and intensive human activities in these urban areas are fueling the problem by increasing emission of pollutants.

In Indian cities, the air pollutants are either emitted from natural sources such as long-range transport of desert dust influx originated from the western arid regions of Africa, Middle-East, and Thar (Rajasthan) regions, predominately during summer and pre-monsoon season (El-Askary et al., 2006) or they can be of anthropogenic origin as given in **Figure 1** (Ghose et al., 2005; Habib et al., 2006; Prasad and Singh, 2007; Badarinath et al., 2010; Sharma et al., 2010; Kharol et al., 2011; Dandotiya et al., 2020).

Vehicular emissions (95%) have been identified as prevalent source of high NO₂ concentrations followed by industries and fuel burning, thereby increasing air pollution in urban areas of India (Mondal et al., 2000; Ghose et al., 2004; ARAI, 2010). The combustion of low-quality fuel in Indian cities causes SO₂ emissions (Zou et al., 2007). Air pollutants are also emitted from crude oil wells and flared natural gas (Amakiri et al., 2009). Besides, open burning and landfill fires of municipal solid waste were recognized as chief source of air pollution for Mumbai, India in a study reported by National Environmental Engineering Research Institute (Central Pollution Control Board (CPCB), 2010). Open coal liming, fluoride mining, lime stone mining, thermal power plant, natural and domestic burning of coal, cement industry, and road dust were another primary source of air pollution in India (MPCB, 2010; Maji et al., 2016). Road dust (61%) was identified as major source of high particulate matter concentration followed by vehicular emissions, industrial emissions, vegetation, and solid fuel burning for another Indian city (Pune). Plastic industry, domestic waste burning, and food processing factories were the main sources of air pollution in Nashik city (Maji et al., 2016). Diesel generators, coal based industrial emissions, oil refinery emissions were major source of PM in Agra (Maji et al., 2016). Also, thermal power plants in most of the cities in addition to large- and small-scale industries are contributing to high air pollution levels. For Kolkata, 51.4% of the air pollution is contributed by motor vehicles followed by 24.5% emissions from industries and 21.1% dust particles (West Bengal Pollution Control Board, 2005).

The deterioration of air quality has been further aggravated by emission of toxic pollutants such as particulate matter, greenhouse gases like SOx, NOx, and O_3 (Rumana et al., 2014). Emission of aerosols from deserts, oceans, forest fires, and volcanoes into the atmosphere also adds to air quality depletion. Increase in population, urbanization, and industrialization has depleted air quality and hence adversely affects human health (Rumana et al., 2014).

Besides, Particulate matter, Black carbon (BC) has been studied by various workers around the globe. Singh A. et al. (2014) reported mass concentrations of BC in Indo-Gangetic Plains (IGP) that varied from 8.5 to 19.6, 2.4 to 18.2, and



2.2 to 9.4 μ g m⁻³ during paddy-residue burning emission in the month of October-November, emission from bio- and fossil-fuel combustion during December–March months and wheat-residue burning emissions duringApril-May, respectively. In contrast, the mass concentrations of Elemental Carbon (EC) varied from 3.8 to 17.5, 2.3 to 8.9, and 2.0 to 8.8 μ g m–3 during these emissions, respectively. Not only this, polycyclic aromatic hydrocarbons (PAHs) have been studied by Rajput et al. (2011) during paddy and wheat biomass burning emissions of Indo-Gangetic plains and reported 40 ng m⁻³ of total PAHs are reported from paddy residue burning and 7 ngm⁻³ during wheat burning season.

Human Health Impacts by Air Pollution

Since last few decades, there has been signification degradation of air quality in most of the Indian cities as many of the Indian cities are in grip of serious air pollution issue such as Kolkata, Delhi (Ghose et al., 2005) with the air quality above the standards provided by CPCB and WHO. The daily and annual average values were high for most of the gaseous pollutants in Indian cities (Dandotiya et al., 2020). The literature suggests the high load of ambient air pollution (specifically in the Indo-Gangetic plain) (Satheesh et al., 2002; Kharol et al., 2011; Ramachandran et al., 2012) has been identified as one of the important contributors to the air pollution related diseases burden in India (Prabhakaran et al., 2020). Rajput et al. (2016) assessed temporal variability and source contributions of PM1, trace metals, five major elements, and four water-soluble inorganic species (WSIS) in the Indo-Gangetic Plains (IGP). Total WSIS contributes about 26% to PM1 mass concentration. Secondary aerosols (contributing as high as) were predominantly derived from stationary combustion sources and contributed ${\sim}60\%$ to PM_1 loading. Further, atmospheric fog prevalent during wintertime can have a severe impact on atmospheric chemistry in the air-shed of IGP.

The health issues linked with air pollution is a topic of major concern specially for developing Indian cities. In India, the outdoor air pollution has become fifth eminent reason of death after high blood pressure, indoor air pollution, poor nutrition and tobacco smoking in 2012 causing about 0.62 million premature excess number of death cases (NYT, 2014; Maji et al., 2016).

Air pollution is linked with short term, medium, or long term impacts on human health (Gumashta and Bijlwan, 2020). Several studies have been conducted regarding the short-term health effects of exposure to air pollution. Short term impacts include irritation to eyes, throat, and nose, and some respiratory infections such as pneumonia and bronchitis, while long term air pollution impacts involve chronic respiratory diseases, heart related problems, lung cancer, and even damage to the brain, liver, kidneys, or nerves (Faheem et al., 2021). Meanwhile Prabhakaran et al. (2020) reported that both short- and long-term exposure to air pollutants contributed to higher blood pressure and increased risk of incident hypertension. The associations between gaseous pollutants and health outcomes have also been discussed (Samoli et al., 2008, 2013; Stafoggia et al., 2013). The higher gaseous and particulate matter concentrations in air are significantly connected with premature mortality and hospitalizations for respiratory and post respiratory illnesses in cities (Burnett et al., 1997; Yang et al., 2004). Rajput et al. (2019) reported that coarse particles exhibited higher mass deposition fraction in extrathoracic region, whereas fine particles deposited
significantly in pulmonary region. Intensification of biomass and biofuel burning emissions during post-monsoon and wintertime have implications to deeper penetration and higher mass deposition fraction of fine-particles in the PUL region.

The air pollution impacts are different in different people such as some individuals are more sensitive to air pollutants than others. Children, elderly people and pregnant women are more prone to health risks related with air pollutants. The studies revealed that the children on exposure to air pollution are highly affected as compared to adults as the lungs of children are comparatively less developed at birth and are not proper functional until about 6–8 years of age (Burri, 1984; Lee, 2010; Smith, 2013). Also, the people who are already suffering from health issues like heart, lung disease, asthma etc are having higher probability to suffer more. Moreover, the extent of impacts depends on duration of exposure and concentration of air pollutants as studied in the city of Agra (Faheem et al., 2021).

Various epidemiological studies conducted in this concern states that poor quality of air poses significant risk to human health creating problems such as decreased lung function, respiratory symptoms and increased asthma incidence, allergy, and cardio-respiratory illness [Ghose et al., 2005; Pope et al., 2009; Beckerman et al., 2012; Portnov et al., 2012; WHO, 2013; Cheng et al., 2014; Tsai and Yang, 2014; Carosino et al., 2015; Global Initiative for Asthma (GINA), 2015; Shaw and Gorai, 2020] with higher concentration of air pollutants. Chronic obstructive pulmonary diseases, influenza, bronchitis, asthma, upper track respiratory infection, and acute respiratory infections were some other health impacts observed for Indian cities due to air pollution (Haque and Singh, 2017). Further, air pollution is linked with several disease and even premature death. The air pollutants dispersed to a long distance and they react or damage the mechanisms by chemical reaction with the molecules of respiratory system and bringing about adverse chemical changes. The health issues such as genetic changes, impaired liver function, hematological abnormalities, and neurobehavioral problems were also associated with air pollution especially for the people exposed to higher vehicular emissions. These include traffic policeman, auto, and taxi drivers and roadside hawkers (Mukhopadhyay, 2009). Besides, detrimental health effects, such as lung cancer, cardiovascular disease risk, cardiopulmonary mortality, and pulmonary inflammation have been reported on exposure to particulate matter in several epidemiological studies (Pope et al., 2009; Huang et al., 2012; Gorai et al., 2014; Prabhakaran et al., 2020).

Furthermore, the health impacts of NO₂ involve irritation of the alveoli and resistance in airways and pulmonary function and decrease in pulmonary capacity reported for cities such as Agra and Taiwan (Mudgal et al., 2000; Yang et al., 2005; Saini et al., 2008). Respiratory health effects, lower birth rates, lower birth weights, and chronic bronchitis are health impacts associated with exposure to high SO₂ concentrations (Ciccone et al., 1995; Dejmek et al., 2000; Rogers et al., 2000). Although gaseous air pollutants such as NO₂ and SO₂ are matter of increasing concern for human health; but particulate matter was observed as prominent reason for air pollution related mortality and morbidity rather than gaseous pollutants (Maji et al., 2016). High particulate matter concentration was observed in India with more than 50% of the population exposed to these higher concentrations (above NAAQs) (Ramya et al., 2021). The premature death due to SPM is reported to be very high and the children are the worst effected groups in Kolkata (Haque and Singh, 2017). Besides, abundance and variability of viable bioaerosols was reported by Rajput et al. (2017) in Indo-Gangetic Plains with very high concentration of Grampositive bacteria (GPB), Gram-negative bacteria (GNB), and Fungi; having implications for human health.

The monitoring of ambient air quality for selected cities in India is conducted by The Central Pollution Control Board (CPCB). In 1984, CPCB initiated National Ambient Air Quality Monitoring (NAAQM) for monitoring air quality that was later renamed as National Air Monitoring Programme (NAMP). Also, the Government of India has prescribed The National Ambient Air Quality Standards (NAAQS). Health risk assessment has been conducted in various studies using formulae given by USEPA.

About 91% of world's population has been found to be residing in areas with air quality higher than permissible limits according to WHO (Mostafavi et al., 2021). Air pollution has led to death of about 3.8 million people throughout the globe as reported by WHO, due to certain human health issues such as ischemic heart disease (27%), pneumonia (27%), chronic obstructive pulmonary disease (20%), stroke (18%), and lung cancer (8%) (Ramya et al., 2021). In India, about 1.24 million deaths have occurred due to air pollution. Out of this, 0.67 million cases were attributed to ambient air pollution and remaining 0.48 million cases were linked with household air pollution (Rumana et al., 2014; Balakrishnan et al., 2019). In India, about 0.62 million in 2005 and 0.69 million in 2010 premature death cases have occurred due to outdoor air pollution (OECD, 2014).

About 1.67 million (95% uncertainty interval) deaths were attributable to air pollution in India in 2019, accounting for 17.8% (15.8–19.5) of the total deaths in the nation. The majority of these deaths were due to ambient particulate matter pollution (0.98 million) and household air pollution (0.61 million). There was a decrease in death rate due to household air pollution by 64.2% from 1990 to 2019, whereas an increase was observed in death rate due to ambient particulate matter pollution by 115.3 and 139.2% increase in death rate due to ambient ozone pollution (GBD 2015 Risk Factors Collaborators).

Air pollution has led to respiratory diseases in about 70% of people in an Indian city, Kolkata as reported jointly by Chittaranjan National Cancer Institute, West Bengal Department of Environment and the Central Pollution Control Board (CPCB) (Mukhopadhyay, 2009). About 10,647 premature deaths were caused due to air pollution in Kolkata in 1995 (Ghose, 2002; Schwela et al., 2006). Adverse lung diseases and genetic abnormalities in exposed lung tissues were reported for children exposed to polluted air in Kolkata (Lahiri et al., 2000). The people residing in Kolkata city were facing seven times higher burden on their lungs due to air pollution as compared to their rural counterparts and about 47% of Kolkata's residents were suffering from lower respiratory tract symptoms Roy et al., 2001; West Bengal Pollution Control Board (WBPCB), 2003; Schwela et al., 2006. Rajeev et al. (2018) reported

health risk assessment of PM1-bound carcinogenic hexavalent chromium [Cr (VI)] from central part of Indo-Gangetic plain (IGP) by assessing excess cancer risk (ECR) which was found to be 57 and 14.3 (in one million) for adults and children, respectively.

The human health impacts caused by air pollution result in high economic cost of about USD 80 billion in 2010, that is almost equal to 5.7% of India's gross domestic product (GDP) (Maji et al., 2016).

Various studies have been conducted regarding air pollution and their associated health impacts for Indian cities such as for Delhi (Gurjar et al., 2010; HEI, 2011; Rizwan et al., 2013; Nagpure et al., 2014); Chandigarh (Gupta et al., 2001); Kolkata (Ghose et al., 2005; Gurjar et al., 2016; Haque and Singh, 2017); Rajasthan (Rumana et al., 2014); Lucknow (Lawrence and Fatima, 2014); Mumbai (Joseph et al., 2003; Maji et al., 2016); Maharashtra (Maji et al., 2016), Agra (Maji et al., 2017); Gwalior City (Dandotiya et al., 2020); Chennai (Jayanthi and Krishnamoorthy, 2006; HEI, 2011). Agarwal et al. (2018) studied mutagenicity and cytotoxicity of PM from biodiesel-fueled engines that were relatively higher compared to their diesel counterparts, indicating the need for exhaust gas after-treatment. The exhaust of chemical characterization revealed that biodiesel-fueled engines contained several harmful PAHs and trace metals, which affected the biological activity of PM.

Aerosols and Particulate Matter

Aerosols have been considered as one of the key air pollutants that significantly influence the air quality and affects public health (Xu et al., 2014). Aerosol optical depth (AOD), an optical property, have been determined in several studies using either ground based observations or satellite data to monitor the concentration of aerosols in the atmosphere. The AOD values are extensively used to represent air pollution level, reflect atmospheric conditions and define climatic effects as these values are closely linked with air pollutants such as PM_{2.5}, PM₁₀, NO₂, SO₂, and O₃ (Chu et al., 2003; Xu et al., 2014; Li et al., 2016; Awais et al., 2018; Ahmad et al., 2020). The monitoring of aerosols has been carried out in many of the Indian cities, as India has been recognized as one of the regional hot spots of aerosols because of increasing anthropogenic activities in the country. The high aerosols load in the atmosphere causes adverse human health impacts and reduces visibility due to poor air quality (Davidson et al., 2005). The exposure to particulate matter (especially PM_{2.5} i.e., particles $<2.5 \,\mu$ m in diameter) has been recognized as fifth leading risk factor throughout the globe and the third leading risk factor in India with about 1 million premature mortality per year across the nation (Chowdhury and Dey, 2016; GBD 2015 Risk Factors Collaborators, 2016; Conibear et al., 2018; Chen et al., 2020). The PM_{2.5} can penetrate deep into the human body and hence can cause greater risk among all other air pollutants (Xing et al., 2016). In India, industrial and vehicular emissions, dust, emissions from biomass burning, open waste burning, household and power sector are major sources of high PM2.5 concentrations (Guo et al., 2017; Conibear et al., 2018; Venkataraman et al., 2018). Domestic cooking and heating, dust from construction activities and industrial emissions are major urban sources of $PM_{2.5}$ (Guttikunda et al., 2019). Rajput et al. (2014) reported the PM2.5 mass concentrations in Patiala region of Punjab during paddy-residue burning in the months of October and November in the range of 60–390 μ gm⁻³ with organic carbon (OC \approx 33%) contributing significantly; while, mass concentration of PM_{2.5} during wheat-residue burning period of April–May varies from 18 to 123 μ gm⁻³.

Besides, emissions in the surrounding rural areas, also contribute to the urban pollution in India (Guttikunda et al., 2019; Ravindra et al., 2019), such as local sources (like traffic, power plants, industries) account for \sim 70% of total PM_{2.5}, but the non-local sources (agricultural crop burning in the neighboring states) contribute over 30% in Delhi (Guo et al., 2017; Prabhakaran et al., 2020). Moreover, the burning of firecrackers during Diwali festival in Delhi worsens the situation by adding more pollutants (Ganguly et al., 2019). With the ongoing urbanization, PM_{2.5} pollution is expected to further increase in the coming decades (Chowdhury et al., 2018; Conibear et al., 2018).

The important studies conducted over Indian cities regarding aerosols and particulate matter are discussed in this section and average AOD values for some of the Indian cities are given in **Table 1**.

Kaskaoutis et al. (2012) conducted a decadal study (2001–2010) for analysis of variations and trends in aerosol properties over Kanpur, India using AERONET data. The study showed overall increase in column-integrated AOD on a yearly basis with significant increase in AOD during the months of November and December as well as for the months of March and April. The increase has been attributed to continuous increase in anthropogenic emissions during which are primarily due to fossil-fuel and biomass combustion over the IGP. Choudhary et al. (2021) studied the seasonal and spatial variability of Brown Carbon (BrC) and reported that water soluble organic carbon (WSOC) aerosols during winter exhibited \sim 1.6 times higher light absorption capacity than in the monsoon season at Kanpur, a central site in Indo-Gangetic Plains.

Further, the aerosols optical properties have been examined during the period 2010–2012 for Greater Noida, Delhi region, using ground-based sun photometer data by Sharma et al. (2014).

In a study conducted for Varanasi, India by Murari V. et al. (2015), the annual mean concentration of particulate matter (PM_{2.5} and PM₁₀) was higher than annual permissible limit (PM₁₀: 80%; PM_{2.5}: 84%) in a range of 8-9 times over than the approved standard values. The study states that high PM values pose a risk of developing cardiovascular and respiratory diseases as well as lung cancer. Further, Sahu and Kota (2017) showed 0.69% increase in non-accidental mortality per 10 μ gm⁻³ increase of PM_{2.5} over Delhi in a study conducted during 2011-2014. Rajeev et al. (2016) attempted to characterize finemode ambient aerosols, and individual rain waters during the South-west monsoon (July-September 2015) in the central Indo-Gangetic Plain (IGP). Not only this, water-soluble ionic species (WSIS) were measured and characteristic mass ratios suggested that below-cloud scavenging was predominant mechanism of aerosols wash-out.

 TABLE 1 | PM values for some Indian cities during different years.

Study area	PM values (μg/m³)	References	
lorhat, Northeast India	24 h mean concentration $PM_{2.5} = 121 \pm 49 \\ PM_{10} = 153 \pm 45$		
Delhi (ITO)	24-h mean concentration $PM_{2.5} = 71.9$	Shaw and Gorai, 2020	
Banglore (S. G. Halli)	24-h mean concentration PM $_{10} = 11.90$		
olkata	24-h mean concentration PM $_{10} = 97.00$		
hubaneswar	Annual mean concentration $PM_{2.5} = 30.6 \pm 22.1$ $PM_{10} = 88.3 \pm 30.6$	Mahapatra et al., 2018	
lanpur	PM_1 average mass concentration During non-foggy conditions = 247 ± 113 During foggy conditions = 107 ± 58	Rajput et al., 2018	
Patiala	$PM_{2.5}$ mass concentration= 60-390 (October–November) $PM_{2.5}$ mass concentration = 18–123 (April–May)	Rajput et al., 2016	
atiala	Average concentration $PM_{2.5} = 55.4 \pm 13.5$	Sen et al., 2016	
ucknow	Average concentration $PM_{2.5} = 51.5 \pm 17.7$ $PM_{10} = 182.2 \pm 58.0$		
(olkata	Average concentration $PM_{2.5} = 47.6 \pm 9.3$ $PM_{10} = 66.7 \pm 17.0$		
lew Delhi	Average concentration $PM_{2.5} = 61.8 \pm 18.6$ $PM_{10} = 127.4 \pm 62.2$		
lagpur	Average concentration $PM_{2.5} = 35.2 \pm 18.4 \\ PM_{10} = 53.9 \pm 23.7$		
/aranasi	Average concentration $PM_{2.5} = 52.5 \pm 28.6$ $PM_{10} = 139.6 \pm 68.0$		
1id-IGP region	Annual mean PM_{10} concentration = 206.2 \pm 77.4	Sharma et al., 2016	
/aranasi	Annual mean concentration $PM_{2.5} = 100.0 \pm 29.6$ $PM_{10} = 176.1 \pm 85.0$ Monthly average concentration $PM_{10} = 43.6-318.5 \ \mu g/m3$ $PM_{2.5} = 50.1-154.0 \ \mu g/m3$	Murari V. et al., 2015	
Delhi	Mean mass concentrations $PM_{2.5} = 118.3 \pm 81.7$ $PM_{10} = 232.1 \pm 131.1$	Tiwari et al., 2015	
ishakhapatnam	Annual average concentration $PM_{10} = 70.4 \pm 29.7$	Guttikunda et al., 2015	
hennai	Annual average concentration $PM_{10} = 121.5 \pm 45.5$		
lelhi	$\label{eq:PM2.5} \begin{array}{l} PM_{2.5} = 130.0 \pm 103.0 \\ PM_{10} = 222.0 \pm 142.0 \end{array}$ Tiwari et al., 2014		
lune	Average mass concentrationYadav and Satsangi, 2013 $PM_{2.5=}$ 72.3 \pm 31.3 $PM_{10} = 113.8 \pm 51.6$		
arapani, foot–hills of NE–Himalaya	Wintertime concentration of $PM_{2.5} = 39-348$	Rajput et al., 2013	
Raipur	Annual average concentrations (July 2009 to June 2010) $PM2.5 = 150.9 \pm 78.6$ $PM10 = 270.5 \pm 105.5$ $PM1 = 72.5 \pm 39.0$	Deshmukh et al., 2013	

(Continued)

TABLE 1 | Continued

Study area	PM values (μg/m³)	References
Mumbai	Average concentration $PM_{2.5} = 42$	Kothai et al., 2011
Agra	PM 2.5 = 104.9 PM10 = 154.2	Kulshrestha et al., 2009
Kolkata	$PM_{10} = 140$	Karar and Gupta, 2006

Adding to PM studies, Singh V. et al. (2021) analyzed particulate matter ($PM_{2.5}$) in five Indian megacities (Chennai, Kolkata, New Delhi, Hyderabad and Mumbai) for 6 years period (2014–2019). Among all cities, Delhi is found to be the most polluted city followed by Kolkata, Mumbai, Hyderabad, and Chennai. Chakraborty et al. (2017) reported high levels of watersoluble organic aerosols (WSOA) and total organic aerosols (OA) using Aerosol Mass Spectrometer in two cities of Indo-Gangetic Plains.

In addition, Chen et al. (2020) discussed the long-term and short-term effects of PM2,5 over four Indian megacities (Delhi, Chennai, Hyderabad and Mumbai) during 2015-2018. The results depict annual averaged PM2.5 concentration of 110 µg/m³ (Delhi), 60 µg/m³ (Mumbai), 56 µg/m³ (Hyderabad), and 33 µg/m³ (Chennai) during study period with worst air quality for Delhi. The study showed 75% increase in PM2.5 concentration during Diwali due to burning of firecrackers that causes 20 extra daily mortality. The long-term exposure to PM2.5 causes 17,200-39,400 premature mortality and 428,900-935,200 years of life lost each year in these four Indian cities. About 10,200, 2,800, 5,200, and 9,500 premature deaths occur each year in Delhi, Chennai, Hyderabad, and Mumbai, respectively, on long-term ambient PM_{2.5} exposures. Among the major diseases, cardiovascular diseases were dominant with ischaemic heart disease (IHD) contributing about 40% and cerebrovascular disease contributing about 30% in each city.

Dutta and Jinsart, 2020 analyzed the PM concentration over Guwahati city during three-year period (2016–2018) and observed high PM levels (>100 μ g m⁻³) during winter season causing high air pollution. The study showed acute health risk to city residents during winter as analyzed from computed hazard quotients (>1). Sorathia et al. (2018) reported diurnal variability of Dicarboxylic Acids (DCAs) and levoglucosan in PM10 during winter over IGP indicating biomass burning emission and secondary transformations to be predominant sources of DCA during wintertime.

Meanwhile, Delhi has been recognized as one of the most heavily polluted cities of India suffering from air pollution caused by industrial and vehicular emissions, thereby possessing high levels of anthropogenic aerosols (Mishra et al., 2013; Singh B. P. et al., 2014). The dust aerosols during pre-monsoon period further worsens the air quality, reduces visibility and increases radiative forcing (Singh et al., 2005, 2010). According to urban air database by WHO in September 2011, high PM 10 (above permissible limits) was observed in Delhi. The high particulate matter concentration causes several respiratory issues that may lead to chronic diseases in Indian cities (Jayaraman, 2007). Tiwari et al. (2009) also reported that $PM_{2.5}$ concentration (97 \pm 56 $\mu g~m^{-3}$) was nine times higher than the air quality guidelines given by World Health Organization (WHO) (2005) over Delhi in 2007.

Besides, particulate matter concentration for Indian megacities during 2010 and 2016 has been discussed in **Figure 2** depicting % increase in almost all the Indian megacities during 6 year period in cities such as Varanasi (Singh et al., 2017; Kumar A. et al., 2020). The increasing PM concentration is correlated with the rising urban population (Kumar P. et al., 2020).

Meanwhile, toxicological studies have established that the toxic effects of particulate matter arise from combined effects of PM size and chemical composition. The modifications in PM composition due to several factors also impart changes in health effects (Peng et al., 2005). These results suggesting that besides mass concentration, chemical composition of PM is also important in evaluation of toxicity on exposure to PM, were supported by Oeder et al. (2012), Kelly and Fussell (2012), and Mirowsky et al. (2013). Further, the source apportionment of aerosols during wintertime has been looked into by various researchers. Rajput et al. (2018) studied secondary formation processes, fog-processing and source-apportionment of PM1bound species in IGP and reported that the foggy conditions were associated with higher contribution of PM1-bound organic matter alongwith approximately equal decrease in SO_4^{2-} , NO_3^{-} , and NH_4^+ and mineral dust fractions.

COVID-19 and Air Quality

Besides the deteriorating air pollution conditions, the improvement in air quality has been reported globally during lockdown imposed due to COVID-19. The restrictions lead to reduction in anthropogenic emissions and hence decrease in PM and gaseous concentrations in most of the cities throughout the globe (Adams, 2020; Berman and Ebisu, 2020; Menut et al., 2020). The similar trend was observed in Indian cities such as for Delhi, Kolkata (Bera et al., 2020; Mahato et al., 2020; Sharma et al., 2020; Singh and Chauhan, 2020; Srivastava et al., 2020; Maji et al., 2021). According to the data provided by NASA, there has been 30% reduction in global NO₂ emissions with 70% decrease in NO₂ emissions in India (Gautam, 2020).

A lockdown period study was conducted by Vadrevu et al. (2020) for analysis of spatio-temporal variations of air pollution (Singh B. P. et al., 2014) (using NO₂ and AOD) for 41 Indian cities. The study revealed about 13% reduction in NO₂ levels during the lockdown as compared to pre-lockdown period. The



 NO_2 levels were reduced by 19% as compared to same duration of previous year. Further, Siddiqui et al. (2020) found 27% improvement in air quality index over 8 five million plus cities of India with an average decrease of 46% in NO_2 levels. The closures of industrial and construction activities during lockdown were reason for improved air quality.

Adding to the research in this field, Srivastava et al. (2020) conducted air pollution study over Lucknow and New Delhi during 21-day lockdown in India by analyzing available data for primary air pollutants (PM2.5, NO2, SO2, and CO). Significant decrease in air pollutants with an improvement in air quality was observed for both the Indian cities. Further, Bera et al. (2020) in a study conducted for Kolkata city stated the reduction in air pollutants such as CO, NO₂, and SO₂ along with particulate matter for the study area as shown in Table 2. The decrease in fossil fuel combustion, vehicular and industrial emissions contributed to significant reduction in air pollutants (CO, NO₂, and SO₂) levels during Covid-19 lockdown. The study further stated the decrease in biomass burning, construction activities and vehicular movement contributed to about 17.5% decrease in PM concentration during Covid-19 lockdown. The improvements in air quality with 30-40% reduction in CO₂ levels with significant temporal variation were observed for Kolkata also by Mitra et al. (2020).

Moreover, the improvement in air quality (PM_{2.5}, NO₂, and AQI) during Covid-19 lockdown was reported by Karuppasamy et al. (2020) revealing improved mortality rates with less number of deaths in India and worldwide due to air pollution. Further, Kant et al. (2020) analyzed decrease in AOD levels during the COVID-19 lockdown period for Eastern Indo-Gangetic planes, peninsular India and North India. On comparison of PM_{2.5} levels over five Indian cities, Kumar A. et al. (2020) observed 50% reduction in PM_{2.5} concentrations over five Indian cities during this period. Goel et al. (2020) in a study conducted for Ludhiana city of India revealed decline in PM_{2.5}, PM₁₀, NH₃, SO₂ concentrations with an overall improvement in air quality index.

TABLE 2 | Percent reduction in air pollutants for Indian cities during COVID-19 lockdown.

Parameter	Location	% reduction	References
NO ₂	New Delhi	61.74%	Vadrevu et al., 2020
	Delhi	60.37%	
	Bangalore	48.25%	
PM _{2.5} , PM ₁₀	Ahmedabad	46.20%	Bera et al., 2020
	Nagpur	46.13%	
	Gandhinagar	45.64%	
	Mumbai	43.08%	
	Kolkata	17.5%	
AOD	Eastern Indo-Gangetic planes, peninsular India and North India	20–37%	Kant et al., 2020
PM _{2.5}	Ghaziabad	85.1%	Lokhandwala and Gautam, 2020
PM _{2.5}	Delhi	35%	Chauhan and Singh 2020
	Mumbai	14%	

Meanwhile, Delhi, the most polluted megacities of India was focused in many of the lockdown period studies. Gupta et al. (2020) reported decrease in CO, SO₂, and ozone levels over Delhi that was supported by significant improvement in air quality over Delhi reported by Kotnala et al. (2020). Significant decrease in PM concentrations was observed for Delhi even in the initial days of lockdown (Maji et al., 2021). Goel V. et al. (2021) analyzed 78% decrease in black carbon during the lockdown and unlock phases for Delhi as compared to the pre-lockdown period. Mahato et al. (2020) also discussed the reduction in the concentration of seven pollutants (PM₁₀, PM_{2.5}, SO₂, NO₂, CO, O₃, and NH₃ gases) over Delhi during lockdown period. The study revealed more than 50% reduction in PM₁₀ and PM_{2.5} concentrations. About 40–50% improvement in air quality was observed using the data collected from 34 monitoring stations. The reduction in levels of various air pollutants for more Indian cities during COVID-19 lockdown period have been presented in **Table 2**.

Such reduction in air pollutant concentration during COVID-19 lockdown is associated with several health benefits. The study conducted by Goel A. et al. (2021) in this concern over Indian cities stated highest health benefits during phase 1 of the lockdown (initial 21 days) due to least PM_{2.5} concentrations during this period. The average pollutant reduction of 44.6% was observed in Uttar Pradesh and about 58.5% decline in Delhi-NCR as compared with year 2019. The tracheobronchial particle deposition was reduced by 30.14% during lockdown. The mortality reduction of 29.85 per 100,000 persons was observed due to declined PM concentrations during 1st phase of lockdown. Also, the decrease in mortality of 8.01 per 100,000 people was analyzed during phase 1 in comparison with the pre-lockdown period in Ghaziabad.

NEXUS BETWEEN URBANIZATION, CLIMATE CHANGE, AIR POLLUTION AND HUMAN HEALTH

The interactions between urban climate, air pollution, and human health in cities need to be explored. The cities in developing nations like India are facing high pressure due to air pollution and climate change. Limited studies have been performed on the combined effects of weather, climate variability, increased air pollution, and health impacts in India (Agarwal et al., 2006; Karar et al., 2006).

Climate plays a considerable role in spatial and temporal distribution of air pollutants. Greenhouse warming and ozone depletion in stratosphere are vital factors of climate change. Climate change can influence the air pollutant concentration and catalyze the formation of secondary pollutants. Also, the climatic conditions in addition to atmospheric parameters, topography and urban settlements influence the dispersion, accumulation and transformation of pollutants in the atmosphere. The dispersal of these air pollutants may cause respiratory disorders such as emphysema, asthma, allergy problems and chronic bronchitis (D'Amato et al., 2002).

According to the World Health Organisation (WHO) estimation, in past 30 years, the precipitation and warming trends due to anthropogenic climate change had taken 150,000 lives annually. The alterations in climate had caused many prevalent human diseases such as cardiovascular mortality and respiratory illnesses due to heat waves etc.

Besides, the nexus between urbanization, climate change and air pollution lies in a way such that some of the atmospheric pollutants (aerosols) can enhance the climate change because of their direct and in-direct effects (Ramachandran and Cherian, 2008). These air pollutants not only degrade the air quality with certain human health impacts but also have a considerable impact on climate by heating lower and mid troposphere, causing sealand temperature gradients, monsoon circulation, distribution of rainfall solar dimming and cloud microphysics (Lau et al., 2006; Gautam et al., 2010; Sharma et al., 2014) thereby modifying the heat wave frequency, intensity of storms and precipitation patterns. These small sized particles can weaken the UHI effect by up to 1 K under heavily polluted conditions (Wu et al., 2017). So, the increased concentrations of air pollutants (such as aerosols) have an impact on global climate change as the increased air pollution (aerosol load) in the atmosphere is associated with the climate system and hydrological cycle (Ramanathan et al., 2001; Jirak and Cotton, 2006). In addition to this, the indirect effect of aerosols can also be seen on optical properties of clouds. Aerosols can affect the surface energy balance by either scattering or absorbing the incoming solar radiations that may cause surface cooling and atmospheric heating (Kaufman et al., 2002; Wu et al., 2017). This influences the radiation equilibrium of Earth via radiative forcing and chemical perturbations (Rosenfeld et al., 2007; Wang et al., 2009; Zhu et al., 2010; Zhang S. et al., 2016; Zhang W. et al., 2016).

Besides, the atmospheric structure and climate is influenced by concentration of atmospheric pollutants that are emitted by human activities (Fischer et al., 2003; Jaffe and Ray, 2007; Yan et al., 2008).

Commercial and high traffic regions have higher concentration of gaseous pollutants than vegetated areas. Also, the concentration of pollutants varies with the seasons and other atmospheric parameters (Dandotiya et al., 2020). The estimation of pollutant concentration is influenced by atmospheric conditions of that urban area such as temperature, relative humidity and wind speed etc.

The greenhouse gases (GHGs) emissions are estimated mainly by consumption patterns in cities of the developed world that causes climate change. According to IPCC report, \sim 20% of global emissions were attributed by buildings. Further, transportation was estimated to contribute to 13% of GHG emissions (Diarmid Campbell-Lendrum and Corvala). It can be seen that both buildings and transportation are eminent factors of cities. Also, the cities face higher pollution issues than rural areas with higher vegetation due to higher emissions from transportation and fossil fuel burning in highly populated regions with high vehicular traffic (Dandotiya et al., 2019).

Further, it is notable that the urbanization phenomenon plays important role in both climate change and air pollution either directly or indirectly. The increase in air pollutant emissions and their concentration in the atmosphere increases with the urbanization. The urban characteristics, materials used, vegetation, vehicular traffic etc alters the climatic conditions of an urban area thus leading to formation of strong spatial gradients of heat and air pollution. These conditions exacerbate the risks for human health.

The expanding urban areas with inadequate or improper management accompanied by land use land cover changes, deforestation and decrease in vegetation cover and alterations in climate variables can influence or modify urban climate by transformation of natural land surface to impervious surfaces (Balica et al., 2012; Jha et al., 2012). The urban heat island effect by increased urban temperature due to climate change increases the demand of energy requirement for cooling in cities. The air conditioners used for reducing the high temperature in cities in turn emits harmful GHGs that cause urban air pollution. Also, the concentration of certain pollutants, such as ozone, is influenced by atmospheric conditions and tends to be higher on warmer days. Moreover, the higher demand of electricity consumption leads to higher burning of fossil fuels that also increases air pollution. Certain respiratory issues can be caused by UHI effect due to depleted air quality by certain cooling agents (Liu and Zhang, 2011). The city residents also suffer from thermal discomfort due to elevated urban temperature by UHI effect resulting in exacerbation of heat-waves (Ohashi et al., 2007). The UHI effect influences air quality as the differential heating generates mesoscale winds that facilitate pollutant movement and circulation causing urban air pollution issues (Agarwal and Tandon, 2010). So, air pollution and climate change are interlinked with adverse impacts on human health in cities.

CONCLUSION

The present review highlights high air pollution levels over most of the Indian megacities with air pollutant levels lying above the permissible limits. The continuous emissions from both anthropogenic as well as natural sources causing high PM concentration with adverse human health impacts highlight the necessity of continuous monitoring of air pollutants over the Indian subcontinent using measurements and remote sensing satellite data.

The essential information regarding air pollutant levels in different megacities of India, provided in this review can help in design of effective mitigation strategies for each city by analyzing vulnerable regions. The data can facilitate a baseline data for air quality modeling studies to predict air pollution levels for effective preparedness, adaption and mitigations plans in tackling air pollution. The high disease burden and mortality linked with air pollution in Indian cities should be emphasized to effectively control air pollutant concentration throughout the nation. Besides, the results depicting reduction in air pollution during COVID-19 lockdown period suggest adoption of such

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short-time restrictions for pollution mitigation across different cities of India to improve the air quality and thus benefit human health.

Further, as stated in the review, India being a developing country is experiencing adverse human health impacts due to climate change. Indian cities are exposed to extreme weather events such as high precipitation, floods, droughts, heat waves with increased temperatures induced by climate change. The increase in health surveillance for heat waves, floods and for vector-borne diseases linked with climate change can help in combating severe human health impacts in near future in Indian cities. Also, the high population density with ongoing urbanization and industrialization are some of the primary factors to be considered to avoid negative health impacts associated with climate change in India. So, essential mitigation and adaptation strategies are required for current and projected climate change impacts mentioned in the review to avoid myriad human health effects in Indian cities because of climate change.

To conclude, the use of advanced technologies such as satellite data with geospatial techniques can be of great help in monitoring and mapping of spatial-temporal distribution patterns of the air pollution and climate change and associated health impacts. So, while focusing on building smart cities in developing nations like India, proper urban planning and sustainable measures should be taken for sustainable urban environment to avoid adverse health impacts.

AUTHOR CONTRIBUTIONS

RK was involved in review of the chapter and preparation of the manuscript. PP was involved in overall supervision of the manuscript and manuscript review and editing. Both authors contributed to the article and approved the submitted version.

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Inhalation Exposure to Atmospheric Nanoparticles and Its Associated Impacts on Human Health: A Review

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Nanoparticles (NPs) are receiving an increasing attention from many scientific communities due to their strong influence on human health. NPs are an important marker of air pollution caused by a variety of natural and anthropogenic sources. Due to their ultrafine size, they can be suspended in the atmosphere for a long time and can thus travel larger distances and cause several health issues after exposure. A variety of NPs that are found in indoor and outdoor settings cause respiratory and cardiovascular diseases. Exposure to NPs through active and passive smoking and household and occupational subjection is reported with thick septum, shortness of breath, and a high level of interleukin protein and tumour necrosis factor (TNF-α) that cause tumour generation in the exposed population. This comprehensive review summarises NPs' source, exposure, and impact on different organ systems. Respiratory models (experimental and computational) used to determine the particle's deposition, airflow transport, and health impact are also discussed. Further, muco-ciliary escalation and macrophage activity, the body's clearance mechanisms after exposure to NPs, have been mentioned. An in-depth analysis of exposure to NPs through inhalation and their health impact has been provided with detailed insights about oxidative stress, inflammation, genotoxicity, and tumourigenicity. Overall, this review offers scientific evidence and background for researchers working in the field of epidemiology, biochemistry, and toxicological studies with reference to atmospheric nanoparticles.

Keywords: nanoparticles, atmosphere, respiratory system, air quality, human health

INTRODUCTION

Among particles of various sizes present in the atmosphere, NPs are particles with sizes of 1–100 nm in diameter (Kumar et al., 2010; Nemmar et al., 2013). The residence time of particles in the diameter range of 0.1–10 μ m is ~1 week. Coarser particulate matter is removed by settling, while smaller particles can only be removed *via* diffusion and coagulation (Scholl et al., 2012; Slezakova et al., 2013; Bakshi et al., 2015). Moreover, the fine size and higher atmospheric retention time of NPs are the basic reasons behind the difficultly in their removal from the atmosphere, consequently causing human health hazard. Nanoparticles originate from vehicular

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exhaust, combustion reactions, forest fires, and industrial emission; they are also produced in the bodies of insects, plants, and humans (Jeevanandam et al., 2018). The atmospheric concentration of nanoparticles is high in Asian countries due to the high rate of urbanisation, industrialisation, vehicular emission, extreme events (dust storms, volcanic eruption, forest fire, etc.), and episodic events (such as firework activity and crop residue burning) (Nakajima et al., 2007; Saxena et al., 2020; Sonwani et al., 2021). Most of the anthropogenic nanoparticles are made up of carbon, silicon, metal, or metal oxides (Querol et al., 2004). India's coal-fired plants emit more than 110,000 tonnes of particulate matter, 4.3 million tonnes of SO₂, and 1.2 million tonnes of NOx per year (Oliveira et al., 2014).

According to the 2007 report of the Intergovernmental Panel on Climate Change (IPCC), nanoparticles act as precursors to coarser particles through their aggregation during the atmospheric ageing process thereby influencing the global climate and urban visibility. Due to their distinct chemical composition and reactivity, they also alter the atmospheric chemistry and global climate (Slezakova et al., 2013). Moreover, these particles are responsible for global warming (by absorbing solar radiation) and cooling effects (by scattering solar radiation), further affecting the global radiation budget (Buseck and Adachi, 2008). Another striking phenomenon is the production of atmospheric brown clouds (ABCs). A study found out that 70% of the ABCs over South Asia are made up of soot emitted from biomass burning, responsible for the melting of glaciers, climate change, and reducing sunlight, which further impacts agricultural production (Engling and Gelencsér, 2010).

NPs also play a significant role in plants by entering via two pathways, viz. apoplastic and symplastic, and thus interact with the cells and environment mostly using Van Der Waals, electrostatic, and stearic forces (Vera-Reyes et al., 2018). NPsbound metal oxides are found to be one of the harmful agents for plants (Giorgetti, 2019). NPs are also one of the phytotoxic agents for plant species, besides tropospheric ozone, that affect the global food security system. NPs are believed to have adverse effects on human health as they enter via multiple exposure pathways such as dermal, inhalation, and ingestion (Sajid et al., 2015). These exposure pathways are known to have a direct connexion with the incipience of neurodegenerative disorders and defects in apoptosis (Chen et al., 2016). With inhalation exposure being the major pathway, NPs enter the body via the nasal passage where it gets deposited on the olfactory mucosa of the nasal chamber more than in any other part of the respiratory system (Seaton et al., 2010). They cause inflammatory responses and oxidative stress in the lungs, impairing the epithelial cells of the system (Rothen-Rutishauser et al., 2008; Nemmar et al., 2013). Bronchial epithelial cells are known to release interleukin 6, a pro-inflammatory cytokine, on exposure to nano-sized diesel exhaust particles (DEP) (Donaldson et al., 2007). The two most prevailing respiratory diseases, asthma and COPD, currently affecting 300 and 210 million people, respectively, across the world are also influenced by exposure to fine particles (Yhee et al., 2016). Moreover, NPs cause acute and chronic problems ranging from asthma and metal fume fever to fibrosis, carcinogenesis, and other chronic lung maladies (Bakand et al.,

2012). Herr et al. (2003) also confirms that respiratory tract infections such as bronchitis, wheezing, and general health issues (tiredness, shivering, and nausea) are caused due to the presence of bioaerosols like pollen and fungi moulds in the air. In addition, some studies prove that exposure to NPs cause intestinal fibrosis, ulcers in the gastrointestinal tract, and oxidative DNA damage in caco-2 cells of the colon (Gerloff et al., 2009; Sajid et al., 2015). In low- and middle-income countries, around 4-8% of recorded mortality is solely caused by indoor smoke from solid fuel, urban air pollution, and occupational airborne particulates, after tobacco, leading to chronic respiratory diseases (Moitra et al., 2015). According to a 2009 WHO report, in India, 488,200 people die prematurely every year as a result of indoor air pollution, whereas 119,900 die prematurely every year because of outdoor air pollution. The number of deaths are increasing and alarmingly high due to rapid industrialisation, urbanisation, and change in lifestyle activities. The rapid population expansion in developing nations like India also impose a huge burden on the physical and mental health of people, especially those residing in megacities (Sonwani and Kulshreshtha, 2016; Saxena and Sonwani, 2019; Goel et al., 2021). The incidence of respiratory diseases in Delhi is 12 times the national average, and 30% of the population was found suffering from 166 different respiratory disorders (Gurjar et al., 2016). Many in vitro and in vivo models of the airway walls of lungs are used to evaluate the extent and outcome of exposure (Nakajima et al., 2007; Slezakova et al., 2013). Several countries are trying to devise measures that can reduce the adverse impacts of atmospheric pollutants (Slezakova et al., 2013; Saxena and Sonwani, 2020); however, there is a lack of studies on the effects of NPs on human health, especially in Asian countries.

Thus, the present review focuses on NPs' source, exposure, and impact on different human body systems, particularly the respiratory system. It also provides a detailed overview of the appearance of toxicological, genotoxic, and tumourigenic effects in the human body due to exposure to NPs. In particular, this is the first review that offers deep insights about the origin of atmospheric NPs, their transportation, deposition, and toxicity, which leads to hazardous diseases in humans.

SOURCES OF NANOPARTICLES

The sources of the nanoparticles in the atmosphere can be natural and anthropogenic. Details on the sources are described in **Figure 1**.

Natural Sources

About 10% of the total aerosols in the atmosphere are generated by human activity, whereas the remaining 90% of atmospheric aerosols are naturally generated (Pipal et al., 2014; Jeevanandam et al., 2018; Sonwani and Saxena, 2021). Nanoparticles are abundant in nature and are released/produced from a variety of sources, mainly from biogenic emission, sea spray, forest fire, volcanic eruption, landslide, and dust storm. Organic compounds form a large proportion of atmospheric nanoparticles because of their ability to react with clouds and air particles (Riipinen et al., 2012). Both natural and anthropogenic



sources make ambient air rich in VOCs, nitrogen oxides, and primary organic aerosols, which ultimately react to form secondary organic aerosols (SOA) (Tiwari and Saxena, 2021). About 90% of the SOA formation is because of biogenic volatile organic compounds (bVOCs) that constitute about 10–40% of the global organic aerosol mass (Volkamer et al., 2006; Saxena and Kulshrestha, 2016; Sonwani et al., 2016).

The chemical composition of NPs differs significantly from place to place as it depends on the types of local sources and their relative contributions. Very few studies have investigated the contribution of NPs in different ambient atmosphere. A study in Pittsburgh mentioned that NPs are mostly composed of organic carbon (OC) and salts of ammonium and sulphate constituting 45–55% and 35–40% respectively (Kuhn et al., 2005); Another study at two sites in Los Angeles found that the composition of OC ranged from 32 to 69%, EC from 1 to 34%, sulphate from 0 to 24%, and nitrate from 0 to 4% (Sardar et al. (2005). Moreover, a study based in two sites in Helsinki trace elements (Ca, Na, Fe, K, and Zn) in higher fractions as compared with heavy metals (Ni, V, Cu, and Pb) that contribute not more than 1% of the total weight of NPs (Pakkanen et al., 2001). Furthermore, incomplete combustion and geological sources are the other significant sources of nanoparticles production in the atmosphere. Forest fires and volcanic sources are the major combustion-generated sources, whereas volcanic eruption along with earthquakes, glaciation, and dust storms are the significant geological sources (Strambeanu et al., 2015). Volcanic ash plays a crucial role in the global transport of toxic chemical species released with NPs (Ermolin et al., 2018). Ash released during volcanic eruptions has a very complex structure and comprises both solid and liquid particulate matter. With time, its composition changes due to cooling and chemical reactions (Strambeanu et al., 2015). The concentrations of toxic elements (Ni, Zn, Cd, Ag, Sn, Se, Te, Hg, Tl, Pb, and Bi) in volcanic ash NPs are seemingly higher than the total content of these elements in any other samples (Ermolin et al., 2018). The eruption of Krakatoa volcano on 27 August 1883 was a remarkable historic event where the smoke column reached 80 km in height and particles were thrown into the ionosphere. This caused strange optical effects and a decline in the global temperature by 1.5°C for the next 2 years in North America and Europe (Strambeanu et al., 2015). Likewise, the Pinatubo eruption of 1991 (Luzon, Philippines) injected vast quantities

of aerosol particles, SO₂, and other sulphate particles into the atmosphere, causing a net effect of global cooling of \sim 0.5°C and significant ozone depletion (Buseck and Adachi, 2008).

Anthropogenic Sources

Urban areas are identified as the centre of anthropogenic sources for NPs as compared with natural sources (Slezakova et al., 2013; Jeevanandam et al., 2018). Anthropogenic sources are classified as either intentional or unintentional. Unintentional sources include incomplete combustion (automobiles and industries), biomass burning, and the incineration of non-biodegradable wastes. Intentional sources consist of the use of pesticides and fertilisers producing a lot of NPs. Based on origin, NPs are also classified into primary and secondary sources. Primary sources include ore extraction, industrial emissions, energy production, and transport activities. Primary sources can be both stationary and mobile. Stationary sources comprise emissions from thermal power plants, chemical and metallurgical industries, and mining activities. Thermal power plants are one of the largest sources of NPs, especially in metropolises where the number of power plants are higher than in any other region like semi-urban/rural areas (Sonwani et al., 2021). The mobile sources are mostly motor vehicles, ships, submarines, aeroplanes, engines, and rockets launched into the extra-atmospheric space (Strambeanu et al., 2015).

Vehicular exhaust is a major source of NPs, released after the incomplete combustion of fuel. It is amongst the leading causes for air pollution due to the rapid rise in vehicular fleet (Walsh, 2011; Banerjee and Christian, 2018). The global vehicle population exceeded 1 billion in 2002 and has continued to climb steadily since then (Walsh, 2011). The major constituents of exhaust include carbon monoxide (CO), hydrocarbons (HCs), nitrogen oxides (NO_x), particulate matter (PM), sulphur oxides (SO_x), volatile organic compounds (VOCs), and their secondary by-products (Walsh, 2011; Banerjee and Christian, 2018). Most particles in vehicle exhaust are in the size range of 20-130 nm for diesel engines and 20-60 nm for petrol engines (Buzea et al., 2007). The sulphur content in fuel is an important parameter as it triggers the nucleation mode in PM formation (Chen et al., 2017). These emissions and ultrafine particles pose a serious risk to environmental health as they are known to produce carcinogenic effects (Banerjee and Christian, 2018).

The second major producer of anthropogenic NPs in the atmosphere are the construction and mining sectors. Surface mining and excavation through mine shafts can produce nanoparticles directly, while NPs' indirect production can occur through decantation, sedimentation, and floatation (Strambeanu et al., 2015). Meteorology and demolition methods also affect the concentration of NPs in the atmosphere. Respirable asbestos fibres, lead, glass, wood, and other toxic particles besides dust are found at the sites of demolition, and these particles can rise high up in the air, sometimes even forming dust clouds that can travel several kilometres, affecting nearby regions (Kumar et al., 2013). The inhalation of metal fumes (copper and zinc) and dust (nickel, chromium, and cobalt) causes pulmonary fibrosis and, in severe cases, cancer, which attributes to 15% of occupational hazards (Buzea et al., 2007).

Transboundary Movement of NPs

The transboundary movement of air pollutants also play an important role in increasing the pollution load in a particular area, raising the NPs' level in the atmosphere. Air mass movement from desert and ocean area significantly affects the air quality in remote locations as it carries a variety of minerals and salts along with transported fine particles (Sonwani and Saxena, 2021). For example, African mineral dust is transported to the Caribbean basin, resulting in seasonal disturbance (Buzea and Pacheco, 2017). Likewise, volcanic eruptions in Iceland in 2011 and the forest fires in Indonesia's Sumatra in 1997 caused environmental havoc in Asia, especially in Singapore and Malaysia. The transportation of fine particle emissions from anthropogenic sources have also been reported by several authors across the world, particularly in the South Asian region (Abas et al., 2019; Cheong et al., 2019; Saxena et al., 2020). Crop residue burning (CRB) is also one of the main factors responsible for the air quality degradation in neighbouring states through transboundary movement, where a large fraction of emission from CRB affects the air quality in the Indo-Gangetic Plains (IGP) (Badarinath et al., 2009a; Saxena et al., 2021a). One of the most affected regions in the IGP is Delhi, which is often affected by CRB activities in the neighbouring states of Punjab and Haryana during the kharif season (Sarkar et al., 2018b; Saxena et al., 2021a). Interestingly, Delhi's air quality has also been affected in the Rabi season due to CRB in Haryana, reported in a recent study by Saxena et al. (2021b). At present, threefourths of the crop residue, amounting to \sim 70–80 million tones, is disposed of by burning; therefore, it becomes a major source of NPs in the air and is well-known to aggravate respiratory disorders. The daily newspapers of northern India during October-November published reports on the incidences of thick clouds of smog formed due to crop residue burning by farmers, reducing the visibility that makes the Air Quality Index (AQI) severe (Badarinath et al., 2009b). In Asia, dust storms are more pronounced around the pre-monsoon period (March to early June) due to high temperatures and wind speed as compared with the other seasons. Such episodic events are largely responsible for contributing to NP pollution in downwind locations (Sarkar et al., 2018a). While in winters, low temperature, lower boundary layers, relatively stable atmosphere, and the transportation of air masses from the adjoining areas enhance the concentration of NPs in that particular area. Badarinath et al. (2009a), using multi-satellite data, reported that crop residue burning in the IGP affects the air quality over the south coast and the Arabian Sea coast of India every year with smoke columns as high as 2.5-3.5 kilometres (Pipal et al., 2014; Singh and Kaskaoutis, 2014; Ravindra et al., 2019).

INHALATION EXPOSURE AND THE EFFECT OF NPs ON THE RESPIRATORY SYSTEM

Human exposure to NPs through inhalation produces various respiratory ailments. Ezzati et al. (2004) argued that indoor air



pollution due to domestic fuel combustion acts as the eighthlargest risk factor for the global burden of disease. The indoor nanoparticle pollution in households is mainly due to the smoke from inefficient burning stoves and biomass fuels used most commonly in rural areas. Traditional practises, illiteracy, and unawareness are some of the key problems that inhibit the people of rural areas from shifting to modern chulas and ecofriendly fuels. The most popular fuel used for domestic cooking in rural regions is the unprocessed biomass solid fuel, known to cause 50 times more pollution than gas stoves (Ravindra et al., 2019). According to a study in rural Senegal, West Africa, out of 1,103 Senegalese women, only 1% used propane gas cylinders (combustion pollution-free) as their primary cooking equipment (Hooper et al., 2015). A 10-year long research carried out in 11 villages of Bangladesh reported 946 cardiopulmonary deaths, out of which 884 people used solid fuels and only 62 deaths were reported in gas-supplied households (Alam et al., 2012). A survey done in 15 homes in Xuanwei and Fuyuan, China, demonstrated experimentally how combustion-derived nanoparticles were generated using all kinds of stoves, while they were significantly reduced with the use of chimneys (Hosgood et al., 2012). An experimental study conducted in Uttar Pradesh, India, reported the average concentration of black carbon (an important fraction of fine particles) emissions was $6-20 \ \mu g/m^3$ and 3–1,070 μ g/m³ for an urban and rural kitchen, respectively (Ravindra et al., 2019). Project Surva, carried out in India, advocated for the use of improved cookstoves widely proposed as a black carbon mitigation measure for Indian households

and hence suggested a need for extensive testing and improved machinery for traditional cookstoves (Kar et al., 2012). Such ecofriendly sustainable techniques can reduce the concentration of NPs in the atmosphere.

Cigarette smoking is yet another common reason for exposure to NPs. One cigarette is known to emit almost 8.8×10^9 nanoparticles. Pertaining to the high particle concentrations and the rapid dilution in air, studies on identifying different types of particles present in cigarette smoke are limited. A study in Netherlands experimentally proved how a longer duration of puff and higher concentrations of tar values in one cigarette contribute directly to a large number of NPs (Williams et al., 2013). Adam et al. (2009) demonstrated the increase in NPs in low-ventilation conditions. They also studied how NP concentration was directly proportional to the number of puffs from a cigarette. Studies suggest an increased incidence of cancers, increased body mass index (BMI), and lower lung function in passive smokers than non-smokers (Gordon et al., 2002; Mohammad et al., 2013). Benzene, 1,3butadiene, and particulate-bound 2,5-dimethylfuran (DMF) are human carcinogens that showed increased levels in non-smokers due to cigarette smoke exposure, thereby proving the adverse effects of passive smoking (Gordon et al., 2002). Occupational health hazards due to exposure to NPs are a common problem rising in developing and under-developed nations. Elevated cases of respiratory and cardiovascular disorders have been recorded in traffic officers due to their excessive exposure to vehicular exhaust. A collaborative study done by India and the



United Kingdom in 2017, wherein various medical tests of 532 traffic officers (exposed to atmospheric NPs) were compared with 150 office workers (working in indoor environments), reported 50% increased cases of thick sputum, pain in joints, and shortness of breath in traffic officers (Bajaj et al., 2017). A similar study conducted in Kathmandu, Nepal, observed increased levels of inflammatory interleukins and tumour necrosis factor (TNF- α) in traffic officers (Shakya et al., 2019). NPs released from welding, flour dust, and diesel exhaust have long-term inflammatory and carcinogenic effects. Welding fumes contain large quantities of partially oxidised, ionised, and reactive metallic nanoparticles (Nasterlack et al., 2008). Another study suggested that the exposure to NPs can cause the production of reactive oxygen species (ROS), cytotoxicity, genotoxicity, and stimulation of ToxTracker reporter cell lines (Bajaj et al., 2017). Asbestos fibre (in all its commercial forms) is classified as one of the potential carcinogens by the International Agency of Research on Cancer (IARC) (Donaldson et al., 2011). Tungsten carbidecobalt dust fumes (WC-Co particles) released during mining and drilling showed deposition in keratinocytes (epidermal cells; skin cell), DNA damage, inhibition of DNA repair, and alterations in gene expression and cell function after short-term exposure (Armstead and Li, 2016). Donaldson et al. (2011) determined the bio-persistence of different size fractions of occupational fibres in several human body systems. Longer slender fibres are usually more persistent, thereby causing various health hazards in our body. Figure 2 illustrates the clearance and deposition mechanisms of these fibres in our body and their potential impacts on humans. The detailed impact of nanoparticles from inhalation to disease site is described in Figure 3.

Mechanism of Exposure to NPs

Authors provide detailed insights about the NP's inhalation, deposition, clearance, and impacts on the respiratory system and

related human body systems. A summary of the effects of NPs on various organs and organ systems of the human body are mentioned in **Table 1**.

Inhalation

Human lungs have an internal surface area between 75 and 140 m^2 and about 3,00,106 alveoli (Buzea et al., 2007). Lungs have a direct contact with the outside environment and hence acts as the main gateway for the entry of nanoparticles into the body. They are also considered important and the primary target site to study the effects of nanoparticles (Bakand et al., 2012; Pacurari et al., 2016).

Air is inhaled *via* nasal and oral routes that pass through the pharynx, progresses into the tracheobronchial tree, and finally reaches the alveoli. The transport of particles dependent on the structure of the respiratory airway, which can be described using the Weibel bifurcating tubes' model, classified into the conducting zone and respiratory zone, as already described in detail. Owing to their small size and high retention time, NPs can diffuse and accumulate in the alveolar region and some may pass through the alveolar epithelium and capillary endothelial cells to enter the cardiovascular system and other internal organs (Buzea et al., 2007; Qiao et al., 2015). Furthermore, electron microscopy proves that the penetration of nanoparticles can occur in both the outer and inner cellular compartments, deep inside the cytoplasm and karyoplasm of pulmonary epithelial and mesothelial cells of the lungs (Bakand et al., 2012).

Deposition

The site of NP deposition in the respiratory tract depends on the particle's aerodynamic diameter (Ferreira et al., 2013). Many fibres with larger diameters are mostly deposited at the "saddle points" in the branching respiratory airways. Thus, they are unable to penetrate deep into the respiratory tract (Buzea et al., 2007). For smaller particles, deposition is mostly governed by Brownian movements. Energy filtering transmission electron microscopy (EFTEM) provides the evidence that aerosol NPs of 20 nm size, within 24 h of inhalation, bypass most of the clearance mechanisms and get deposited in the alveolar region of the lungs (Geiser, 2010).

According to the classical model developed by the International Commission on Radiological Protection (ICRP), NPs (≤ 20 nm diameter) have a high probability to reach the alveolar region (Kreyling et al., 2006; Ferreira et al., 2013). Thus, particles larger than 20 nm in diameter are restricted in the previous sections of the respiratory tract through mucociliary escalation and macrophage activity. Such activities help with the clearance of particles in the tract before they enter the pulmonary region.

Clearance

The deposition is chiefly determined by particle size, the ventilatory parameters, and airway characteristics, whereas the clearance of particulate compounds, once deposited, is dependent on the physicochemical characteristics of the compound. During inhalation, larger particles are deposited at the extra-thoracic region (nose and larynx) and intrathoracic

TABLE 1 | Summary of effects of nanoparticles on various organs and organ systems of the human body.

S. No.	Biological system	Target organ	Effect of nanoparticles	References
1.	Respiratory system	Lungs	Inflammation Oxidative-stress Genotoxicity Tumorogenicity	Clift et al., 2011 Sharma et al., 2012 Zhu et al., 2013 Stueckle et al., 2017
		Alveoli	Activation of macrophages causing pro-inflammatory cytokine release	Nho, 2020
2.	Digestive system	Stomach	Increased mucus production and Inflammation	Georgantzopoulou et al., 2015
		Intestine	Accumulation in lamina propria and degenerating goblet cells	Zhang et al., 2014
3.	Cardiovascular system	Heart	Increased blood pressure and decreased heart rate	Yu et al., 2016
			Increased ROS production leading to oxidative stress	Miller et al., 2012
			Heart muscle fibre degeneration and cellular necrosis	Yu et al., 2016
		Endothelial lining	Inhibition of NO pathways and vasoconstriction	Mills and Miller, 2011
4.	Excretory system	Kidneys	Accumulation in Proximal convoluted tubules	Pujalté et al., 2011
			Nephrotoxicity and DNA-Damage	Sramkova et al., 2019
			Shrinkage of kidney cells and nuclear condensation leading to oxidative stress	Wang et al., 2009; Pujalté et al., 2011
			Cytotoxic effects on glomerular and renal cells	Pujalté et al., 2011
		Epithelial lining	Cytotoxcity in HK-2 cell lines of the lining	Pujalté et al., 2011
5.	Neural system	Brain	Accumulation and effecting expressions of genes	Yang et al., 2010
			Apoptosis, cell cycle alterations and oxidative DNA damage	Valdiglesias et al., 2013
			Inflammation and hormone imbalance	Haase et al., 2012; Feng et al., 2015
6.	Reproductive system	Ovary	Decrease in tissue weight and abnormal cell structures	Asadi et al., 2019
		Testes	Inflammation and cytotoxicity	Santonastaso et al., 2019
		Gametes	Loss of integrity of sperm DNA	Santonastaso et al., 2019
			Problems in the process of oogenesis leading to hormone disbalance	lavicoli et al., 2013
		Epithelial lining	ROS-Overproduction, Oxidative stress and Apoptosis	lavicoli et al., 2013
7.	Endocrine system	Thyroid	Overproduction of T-3 Thyroid hormone and decrease in thyroid-stimulating hormone	Jiang et al., 2019
		Hormone receptors	Blockage of signal cascades, Overstimulation of hormones	Leso et al., 2018
		EDCs (Endocrine- disrupting chemicals)	Cytotoxicity and imbalance in production	lavicoli et al., 2013; Lu et al., 2013
		Kidney	Degrading effects on testes and kidney hormones	Li et al., 2009
		Liver	Elevated serum levels of Alanine aminotransferase Alkaline phosphatase 	Miller et al., 2012; Sharma et al., 2012
			Lipid peroxidation and pathological lesion	

bifurcations due to impaction. These upper respiratory airways are covered with mucus (a complex fluid containing mucins [hydrogel-forming glycoproteins] that protect the body from environmental influences) (Moller et al., 2004; Kirch et al., 2012). This mucus layer traps the large particles, which are transported out by ciliary beating, removing the inhaled NPs, making it the dominant pathway of clearance from the regions of trachea, bronchus, and upper bronchioles (Moller et al., 2004; Geiser, 2010; Kirch et al., 2012). The outer viscoelastic mucus blanket has an underlying periciliary layer that provides conditions for an efficient ciliary beating cycle. The tips of the cilia reach the mucus blanket and accelerate this layer by a coordinated beating (Kirch et al., 2012). Upon entering the pharyngeal region, the mucus along with the NPs are finally swallowed and undergo further processing in the gastrointestinal tract (Henning et al., 2010). Smaller particles penetrate deep inside the lungs where air velocity decreases substantially and are deposited in the bronchioles and alveoli. This is the site where



NPs undergo mucus transport and macrophages participate in the phagocytosis of biogenic NPs like viruses, protozoans, and bacteria (Moller et al., 2004). Macrophage-associated clearance is a much slower process as compared with mucociliary clearance (Henning et al., 2010). Most of these NPs translocate to the connective tissues and involve in the blood circulation, bringing the NPs in contact with the macrophage populations in the non-alveolar region. NPs are mostly cleared by interstitial and intravascular macrophages, as surface macrophages are relatively less effective in NP clearance (Geiser, 2010). After the inhalation and deposition of NPs on the surfactant layer with the underlying epithelial lining fluid, they come in contact with several proteins and biomolecules (including opsonins that enhance phagocytosis by marking an antigen), which makes NPs more susceptible to macrophages. It was observed that there was less phagocytosis in the absence of opsonins; moreover, rapid phagocytosis by alveolar macrophages was reported in the case of opsonindirected migration towards the NPs (Kreyling et al., 2006).

Lung Burden

A significant difference is observed in the behaviour and clearance of soluble and insoluble NPs in the lungs. Soluble NPs dissolve in the aqueous fluid and further goes into the cardiovascular systems. In contrast, the insoluble NPs (black carbon) are removed through mucociliary escalator or macrophage phagocytosis (Buzea et al., 2007). It has been reported that the insoluble NPs cause more tissue damage, inflammation, and lung tumours (Ferreira et al., 2013). The insoluble NPs accumulate more rapidly, exceeding

the macrophage clearing capacity, and hence the defence mechanisms of the lungs fail to operate, resulting in lung injury (Buzea et al., 2007). It was also observed that acute lung inflammation is induced by the production of IL-1 β and TGF- β 1 for a shorter period in the bronchoalveolar lavage fluid. However, a longer exposure duration leads to collagen production, causing lung burden and potential pulmonary fibrosis (Lin et al., 2014).

Human health hazards due to lung burden is directly associated with the increasing cases of premature mortality, especially in developing nations. The determination of lung burden is dependent on the rate of particle deposition, rate of clearance, and residence time of the NPs (Buzea et al., 2007). Morphological observations and retrospective evidence indicate a slower particle clearance of insoluble NPs in large mammalian species (humans and primates) and a greater tendency for retained lung burden, which can translocate from the original alveolar deposition sites to the interstitial compartment of the respiratory system (Warheit, 2004). A long-term exposure to NPs causes high lung burden that leads to carcinogenesis in the lung tissues (Buzea et al., 2007). However, this level of exposure rarely happens in humans, suggesting lower chances of acquiring the malady (Donaldson and Poland, 2012).

Nano-Toxicity

Figure 4 explains the impact of toxicity generated through NP inhalation on human body. Human lungs constitute pseudostratified epithelium in the lung-blood stream barrier. The airways are composed of thin columnar epithelium, together with the bronchial epithelium (3–5 mm) and bronchiolar

epithelium (0.5–1 mm), that all protected with mucous layer (Praphawatvet et al., 2020). The lung tissue consists of more than 40 different cells, so to study the aggregated effect of NPs on lungs, different cell models were made and tested. The exposure to NPs primarily affects the epithelial lining of the respiratory system, where A549 cell lines (derived from the human adenocarcinoma of the lung) are most often used in toxicity testing, and Calu-3, 16HBE140-, and BEAS-2B cell lines are used as models for the bronchial barrier system (Fröhlich and Salar-Behzadi, 2014).

The size of NPs is inversely proportional to the damage they cause in the respiratory tract, as smaller particles easily reach and get deposited in the distal parts of the respiratory tract (Donaldson et al., 2005).

Four main types of toxicological effects of nanoparticles are discussed as follows:

- Oxidative Stress
- Inflammation
- Genotoxicity
- Tumourigenicity

Oxidative Stress

Oxidative stress is the prime consequence of nano-toxicity that occurs due to the imbalance between free radicals and antioxidants in the body in such a way that the extra free radicals with uneven number of electrons react inadvertently with other molecules to produce an imbalance in the respiratory system (Sharma et al., 2012).

Reactive Oxygen Species (ROS) Generation. The production of ROS is one of the important oxidative steps for creating toxicity in human lungs. The overproduction of ROS is caused by NPs' activity in the mitochondrial electron transport chain of the cell (Donaldson et al., 2010). Metallic NPs trigger Fenton-type reactions causing free-radical-mediated toxicity, and carbon NPs are known to affect mitochondrial reactions (Martin and Sarkar, 2017). The increased potential of hydrogen peroxide (H₂O₂) (due to special oxygen transfer properties of some metals) causes the generation of highly reactive hydroxyl radicals (OH⁻), and this process is called as Fenton's reaction (Li et al., 2008).

$$\begin{split} Fe^{2+} &+ H_2O_2 - - - > Fe^{3+} + OH \cdot + OH^- \\ Fe^{3+} &+ H_2O_2 - - - > Fe^{2+} + OOH \cdot + H^+ \end{split}$$

The extent of ROS production (produced by a particular NP) depends on the catalytic activity of surface groups. In a particle of 30 nm in size, about 10% of its molecules are expressed, whereas in the case particles of 10 and 3 nm in size, only around 20 and 50% molecules are expressed respectively (Hao and Chen, 2012).

The deposition of nanoparticles leads to the formation of molecular oxygen-dependent superoxide anion radicals (O_2^{2-}) , H_2O_2 , and hydroxyl radicals (OH⁻). These chemical species are proven to have cytotoxic responses in BEAS-2B bronchial epithelial cells. It was also reported that inflammatory phagocytes (macrophages and neutrophils) are also released from the respiratory system in response to NPs. Such inflammatory phagocytes induce oxidative outburst as a defence mechanism

and ultimately raises the number of ROS in that particular region (Martin and Sarkar, 2017).

The Creation of Oxidative Stress. Oxidative stress is caused in the body either due to the overproduction of ROS or by the weakening of the processes of antioxidant defence. Another factor that determines the presence of oxidative stress in a region is the GSH/GSSG ratio, i.e., the cellular glutathione/glutathione disulphide ratio. Low values of this ratio indicate stress and toxicity in the tested region, where stress is created when lower quantities of glutathione are present and glutathione disulphide accumulates (Nel et al., 2006; Hao and Chen, 2012).

An *in vitro* study by scientists in Korea on BEAS-2B cell lines clearly indicated a rise in oxidative stress due to silica nanoparticle exposure as well as the induction of HO-1 (heme oxygenase-1—an antioxidant enzyme) *via* the Nrf-2–ERK MAP kinase signalling pathway, which suggests how oxidative stress disrupts the ROS balance of the cells (Huang et al., 2010). Meanwhile, in another study done in the United States, oxidative stress caused by ZnO NPs was directly linked to the increased calcium levels in cells, causing cellular dysfunction, impaired signal transduction, and other diseased states (Nho, 2020). Scientists have raised concerns over the inadequate results pertaining to NP exposure gained by *in vitro* models and have therefore adapted a more realistic approach by switching to *in vivo* models to study these xenobiotics (Ursini et al., 2014).

A study conducted in China in 2012 on the different organs of carp (Cyprinus carpio), wherein three different concentrations of 0.5, 5, and 50 mg/L of zinc oxide (ZnO) nanoparticles were tested for their effects on the concentrations of antioxidant enzymes (e.g., superoxide dismutase [SOD], catalase [CAT], etc.) before and after administration, indicated an increased mucus production and apoptosis of cells at higher concentrations and exposure time of ZnO NPs (Cho et al., 2012). TiO₂ nanoparticles on a 90-day exposure increased the concentration of ROS as well as the level of lipid peroxidation and therefore decreased the antioxidant capacity of the lungs, leading to the creation of oxidative stress, which is the direct cause of inflammation and genotoxicity (Lu et al., 2014). Oxidative response is directly linked to inflammatory response as it is proven to cause chronic inflammation. The macrophages of the immune system are known to produce free radicals (the cause of oxidative stress) and pro-inflammatory hormones. A mitogen-activated protein kinase (MAPK) pathway activates and produces transcription factors that ultimately produce proteins responsible for inflammation, cell stress, cancer in the cell, etc.

Inflammation

Our immune system is divided into innate and adaptive immune systems. The innate immune system is the first line of defence against any foreign particle encountering on entering the body. If the innate immune system is not able to deactivate the foreign particle (antigen) on its own, it then induces the adaptive immune system, which is much more complex and effective. This activation is carried out by the dendritic cells in the body. Nanoparticles, being foreign particles, also activate dendritic cells, which in turn generate ROS, cytokines, and chemokines and activate naïve T-cells and various inflammasomes.

Determinants of Inflammation. NPs elicit an inflammatory response in the respiratory system as well as the other systems of the body (Medina et al., 2007; Clift et al., 2011; Padmanabhan and Kyriakides, 2015). A well-known determinant of an NP's ability to cause inflammation is the zeta potential (ξP) of the particle. ξP is the electric potential created when charged groups (present at the surface of a particle) interact with the suspension medium. Since the medium in human body is acidic, if more positively charged groups are present on the surface, more will be the solubility of that particle in the medium and hence there will be more interaction with macrophages, leading to inflammation (Schins, 2013). Magnesium and zinc oxide nanoparticles show high solubility in these solutions. A study reported that non-biodegradable NPs and cationic polymers induce even more inflammation than biodegradable and anionic makeshifts as titanium- and siliconbased NPs induce more inflammation than zinc (Nishi et al., 2020).

The presence of white blood cells (WBCs) in blood is another measure used to determine the presence of inflammation in the body. Inflammation is noticed based on the increase in WBC count above the normal range, which signals a decreasing immunity. When NPs enter via inhalation, their deposition in the lungs leads to the release of pro-inflammatory hormones, and they encounter alveolar macrophages (as they constitute the first line of immunity). Thus, the inactive macrophages become activated to stimulate the movement of various types of proinflammatory cytokines (IL-1 family, IL-6, IL-8, and i-CAM proinflammatory protein expression) to the affected site (Clift et al., 2011; Foldbjerg et al., 2011). An experiment was performed in the United Kingdom using eight different metal oxides in an in vivo rat model to investigate the effect of exposure on bronchoalveolar lavage fluid (BALF). This study reported that the cerium and nickel metal oxides are associated with an immediate neutrophilic cytotoxicity pattern, wherein the increased levels of IL-1β, MIP-2, and LDH indicate neutrophilic inflammation, and the same has been observed for copper and zinc by the end of 4 weeks of exposure (Magdolenova et al., 2014). Another study demonstrates that particle size is directly linked to inflammation, and this has been proven by an experiment performed on rats, wherein they were exposed to equal masses of titanium dioxide in two particle size ranges, showing more retention of ultrafine particles in the lung interstitium and the development of an increased inflammatory response (Peters et al., 2007). It was also reported that A549 cells (human alveolar carcinoma cells) were used to detect the change in cytokine secretion after treating the cell lines with TiO2 NPs and an increased release of the pro-inflammatory hormone interleukin-6 (Medina et al., 2007). Moreover, an in vivo experiment with nickel oxide NPs on rats' lungs found that chronic lung inflammations persist for long durations after exposure (Brown et al., 2001). A similar study done by Padmanabhan and Kyriakides (2015) showed that rats exposed to C13 ultrafine particles were deposited more in olfactory bulb on Day 7 than on Day 1, which clearly indicates how these particles get circulated in the body after inhalation.

The Adverse Effects of Inflammation. Inflammation results in the destruction of cilia in the respiratory tract and hence impairs the movement of mucus (mucus basically traps infectious agents and dirt) through cilia. Inflammation also leads to epithelial injury by breaking the epithelial barrier between the lung surface and blood stream, leading to clotting, decreased lung function, the impaired circulation of blood and oxygen in the body. Pneumonitis and the inflammation of lung tissue are the symptoms of lung cancer, pulmonary fibrosis, chronic obstructive pulmonary disease (COPD), asthma, and cystic fibrosis. Various types of pulmonary fibrosis are the main causes of death in patients diagnosed with chronic lung diseases (Chen et al., 2006). This has been proved in a study done on rats, wherein they were exposed to a long multi-walled carbon nanotube for 30 days, which showed the presence of irreversible granuloma, considered as chronic inflammation in lungs (Lu et al., 2014). Inflammation also results in blood clotting, which activates thrombosis cascade at different locations, ultimately leading to problems in the cardiovascular system, upsetting the cardiac rhythm and increasing the chances of cardiac arrest in the body (Donaldson et al., 2005).

Genotoxicity

Genotoxicity can be induced either by the direct interaction of NPs with DNA or indirectly because of oxidative stress or inflammation. Based on different studies and consequent effects, genotoxicity can be divided into primary and secondary genotoxicity. Primary genotoxicity occurs when ultrafine particles enter the nucleus and cause modifications in DNA. Another primary indirect mechanism is related to the DNA repair cascade. Secondary genotoxicity is caused by oxidative stress and inflammation induced by NPs (Zhu et al., 2013; Magdolenova et al., 2014). **Figure 5** shows silica NPs' deposition in the lungs (via p53 signalling) and the related genotoxicity involved in the tumour formation in rats.

Primary Genotoxicity

Direct Primary Genotoxicity

The oxidation of DNA results in mutagenic modifications such as the hydroxylation of adenine and guanine, thereby leading to the formation of DNA adducts (Donaldson et al., 2005; Berube et al., 2007). DNA adducts are a result of the covalent modifications in DNA in the presence of certain carcinogens. Particulate carcinogens such as asbestos, crystalline silica, etc., have the ability to directly enter the cellular membranes and nucleus. They then interact/disrupt the process and functionality of the different components of the mitotic spindle and ergo produces dysfunctionalities (Magdolenova et al., 2014). These foreign NPs can disrupt all cellular processes including mitosis, the replication of DNA, and the transcription of DNA into mRNA. A study reported that aluminium NPs induced structural damage and DNA instability, besides other mechanisms that explained the DNA damage (including single-strand breaks and double-strand breaks), DNA deletions, and genomic instability



in the form of an increase in 8-hydroxy-20 -deoxyguanosine levels (Donaldson et al., 2013; Chakraborty et al., 2018). Cobalt-chromium nanoparticles significantly increased singleand double-strand breaks and the chromosomal aberrations in mice on exposure, consequently affecting the brain cells in the resulting offspring (Clift et al., 2011).

Indirect Primary Genotoxicity

In the indirect method, NPs attach to the nuclear proteins responsible for DNA repair mechanisms, disrupting their functioning and hence indirectly favouring slip-ups in the nuclear processes. Topoisomerases are the enzymes responsible for carrying out this repair cascade. An experiment showed that Carbon-60 fullerenes can bind to human topoisomerase-II alpha in the ATP-binding domain and therefore disturb its enzymatic activity (Donaldson et al., 2013; Magdolenova et al., 2014).

Secondary Genotoxicity

Secondary genotoxicity refers to the errors created in the structure and function of nuclear components due to the increased ROS production and inflammation, ultimately leading to disrupted DNA sequences. Nanoparticles trigger ROS production in phagocytes (macrophages, neutrophils, etc.), which induces DNA damage and mutagenesis in the neighbouring cells (Donaldson et al., 2013; Magdolenova et al., 2014). It has been reported that in any mechanism, silica triggers the production of ROS leading to the activation of NF-kß (necrosis factor) and AP-1 (primarily in gene expression), causing an increase in the quantities of growth factors and oncogenes that causes mutations and ultimately cancer (Hong et al., 2017). Exposure to diesel exhaust particles produces inflammation, leading to epigenetic changes that include impairment in gene expression, the formation of adducts, cell proliferation, and interruptions in DNA repair (Shi et al., 1998). In another experiment, researchers demonstrated how the A549 cell line (present in alveoli) is more susceptible to DNA damage than the BEAS-2B cell line (present in bronchi) using FPG comet assay (Medina et al., 2007). A study undertaken in Denmark on the A549 cell lines clearly demonstrates that silver NPs induce oxidative stress correlated to cytotoxicity and genotoxicity (Berube et al., 2007).

Tumourigenicity

The MAPK/ERK pathway (also known as the Ras-Raf-MEK-ERK pathway) is a chain of proteins in the cell that communicates a signal from a receptor on the cell's surface to the DNA in the cell nucleus. The signal starts when a signalling molecule binds to the receptor on the cell surface called epidermal growth factor receptor (EGFR). In normal state, extracellular ligands such as epidermal growth factors bind to this receptor and phosphorylates activates it, signalling a cascade of docking proteins that ultimately result in the making of mRNA, which gets encoded to form various proteins.

In this case, ROS and sometimes direct NPs interact with these receptors, and abnormal proteins could either be formed or not due to the changes in the DNA code. When one of the proteins in the pathway mutates, it can become stuck in the "on" or "off" position, which is a fundamental step in the development of many cancers. The entry of NPs plays a major role in increasing the levels of 8-hydroxy-2[']-deoxyguanosine (8-OHdG) in the body. 8-OHdG can give rise to G-to-T transversion mutations in the key genes known to be involved in the development of cancer, hence giving rise to tumours (Guo et al., 2017). Thus, an excessive level of 8-OHdG is directly proportional to the occurrence of carcinogenicity that weakens the immunity. A study observed the levels of 8-OHdG in the urine and white blood cells of 130 workers handling indium tin oxide who were easily exposed to metal oxide NPs (Falcone et al., 2018). Other pathways include p53 inactivation and caveolin-1 overexpression. p53 inactivation occurs when NPs decrease the phosphate levels (required to activate proteins) in cells and therefore increase the chances of tumour formation (Stueckle et al., 2017). Likewise, there are many proteins that are found in high concentrations in the body in the presence of a tumour, and such high levels of proteins are often used to test the presence of neoplasm (the abnormal growth of cells). Caveolin-1 protein's overexpression is associated with carcinogenesis and metastasis. A study reported the inhalation exposure to Titanium oxide NPs on mice, wherein an increased level of squamous cell carcinoma antigen (SCC-Ag) (tumour marker) was observed in high concentrations due to the formation of tumours (Shi et al., 1998).

Tumour suppressor genes in the body are responsible for expressing proteins that prevent the uncontrolled growth of cells forming tumours and hence the decrease chances of genomic mutability. Mutated tumour suppressor genes due to the genotoxicity caused by NPs aren't able to carry out their function, eventually leading to the loss of growth regulation (Luanpitpong et al., 2010). Liou et al. (2017) identified the appearance of cancer stem-like cells on chronic exposure to carbon NPs. Ceria NPs have an ability to generate ROS and cause DNA damage, leading to alveolar papillary neoplasm and carcinoma. Carcinoma also depends on particle size, bio-persistence, and the co-morbidity in an individual. Other recent in vivo studies confirm that the iron oxide NPs as an important cause of hyperplasia and fibrosis of the lungs (Manke et al., 2013). Thus, an excessive increase in fibrosis leads to the death of an individual. Chromium, nickel, and iron oxides released with NPs from welding fumes act as major carcinogens for the human body that significantly increase the chances of tumour formation (Chen et al., 2006).



EFFECT OF NANOPARTICLES ON OTHER SYSTEMS

As discussed previously, the respiratory system is one of the most important pathways for the entry of airborne pollutants into the body and the distribution of NPs to the other biological systems of the body. The retained NPs in the lungs come in contact with the alveoli-blood barrier (present between the alveoli of the lungs and the transporting vessels of the circulatory system), which helps in the exchange of gases stored by alveoli to the entire body and vice versa by the process of diffusion of air particles. The retained NPs seep into these vessels through endocytosis in the alveolar epithelial cells and get transported to the other organs via blood. Endocytosis is a process by which a cell transports particles in and out of the cell by the formation of vacuoles. The second mechanism involves in the diffusion of these ultrafine particles into the olfactory bulbs of neurons via the nasal epithelial barrier for reaching the central nervous system of the body. This system is spread throughout the body and therefore the NPs travel and get deposited in distant places of the different systems of the body (Iversen et al., 2011; Elsaesser and Howard, 2012). Figure 6 shows the impact of exposure to NPs on the different systems of the human body.

Effect of Nanoparticles on the Digestive System

The digestive system also called as the gastrointestinal tract consists mainly of the oesophagus, stomach, and intestines. NP

exposure to the gut mostly occurs either through water or food intake. The presence of silver NPs increases the interleukin-8 levels, directly linked to inflammation and increased mucus production (Georgantzopoulou et al., 2015). Depending on their size, some NPs escape the junctions between the intestinal epithelial cells to enter blood vessels, perturbing the mucus and epithelial cell layers. Furthermore, they finally accumulate in the lamina propria of the intestines where they disrupt the function of goblet cells (Zhang et al., 2014). If there are already underlying conditions such as Crohn's disease or ulcerative colitis in the individual, then the rate of NP accumulation is even higher, leading to inflammation in the intestinal areas (Lomer et al., 2004; Jones et al., 2015). Consequently, the toxicity levels get increased and will lead to colon cancer and other carcinomas.

Effect of Nanoparticles on the Cardiovascular System

NPs can enter our body via inhalation (intranasal or intratracheal) or by oral exposure to the gastro-intestinal tract, which subsequently reaches the circulatory system. These can cause significant alterations in the normal functioning of the system. The increase in blood pressure, decrease in heart rate, and altered vascular tone and dysfunction are some of the initial effects of NPs on the cardiovascular system (Yu et al., 2016). Depending upon the physical properties, concentration, and retention time of NPs, they can have angiogenic/antiangiogenic, vasodilation/vasoconstriction, prooxidant/antioxidant, cytotoxic, apoptotic, and phagocytic effects on our body (Gonzalez et al., 2016). Individuals with pre-existing cardiovascular diseases are more susceptible to these changes. Such persons are more prone to sudden cardiac arrests, heart attacks, and blood clotting due to the assimilation of NPs in the blood stream. Epidemiological studies have revealed evidence of both the acute and chronic effects of NPs (Donaldson et al., 2013). An increased ROS generation because of nanoparticles has been observed as the main cause for these maladies due to the increased oxidative stress (Miller et al., 2012).

Silver NPs that are currently used in pacemakers, drugs, and coupled antibodies have a dual and opposite effect on blood composition, angiogenesis (vessel formation), and the permeability of membranes (Gonzalez et al., 2016). Long-term exposure to TiO_2 NPs can lead to their accumulation in the heart, causing sparse cardiac muscle fibres, inflammation, cellular necrosis, and cardiac biochemical dysfunction. A study on SiO₂ NPs in old rats suggested increased Fbg concentrations and blood viscosity along with myocardial ischemic damage and atrioventricular blockage (Yu et al., 2016). Experiments performed on rodents by Mills and Miller (2011) support the aforementioned results and also provide evidence to endothelial cell dysfunction, the inhibition of NO pathways, and the enhanced responsiveness to vasoconstrictors.

Effect of Nanoparticles on the Excretory System

The kidney contains a network of blood capillaries in each nephron that philtres the toxins from blood; therefore, it is quite

susceptible to xenobiotics and the bioaccumulation of toxins (Pujalté et al., 2011). NPs, after glomerular filtration, tend to concentrate in the proximal convoluted tubules (PCT), which can lead to internalisation *via* tubular cells through endocytosis. A study on TH1 cells exposed to inorganic NPs showed not only the acceleration of NP-induced nephrotoxicity but also the DNA damage in these cells (Sramkova et al., 2019). Similar effects were observed by another study when tubular cells in mice were exposed to copper NPs (Pujalté et al., 2011). This will further lead to genetic disorders and chromosomal aberrations in the genetic material.

Wang et al. (2009) observed that the smaller the particle, the greater its toxicity. This study on HEK293 cells (cultured human embryonic kidney cells) demonstrated that kidney cells underwent shrinkage and nuclear condensation, which are signs of apoptosis on exposure to SiO₂ nanoparticles at dosage levels of 20-100 µg/ml. ROS production in HEK293 cells and oxidative stress induction also indicate the nephrotoxic potential by nanoparticles. Impaired nephrotoxic potential causes dysfunction in the elasticity of nephrons and may cause cerebral epilepsy. The cytotoxicity of NPs was also evaluated in the IP15 (glomerular mesangial) and HK-2 (epithelial proximal) cell lines (Pujalté et al., 2011). The study observed that ZnO and CdS nanoparticles exerted cytotoxic effects on glomerular and tubular human renal cells. These effects were correlated with metal composition, particle size, and metal solubility (Pujalté et al., 2011).

Effect of Nanoparticles on the Neural System

Neurons together with neuroglial cells constitute the nervous tissue making up the nervous system. NPs can reach the central nervous system (CNS) via different routes, most commonly by inhalation that can reach the brain through the upper respiratory tract to the olfactory bulb to the trigeminal nerve to the trigeminal nucleus and thalamus to the blood-brain barrier and ultimately to different parts of the brain (Haase et al., 2012; Feng et al., 2015). NPs produce toxicity effects on neural cells by accumulating in different regions of the brain and affecting the expression of genes that helps in the development and function of CNS (Yang et al., 2010). The malfunctioning of CNS can show symptoms like depression, lack of concentration, and even mental retardation. ZnO NPs induce significant cytotoxicity, including a decrease in viability, apoptosis, cell cycle alterations, and different kinds of genetic damage such as oxidative DNA damage (Valdiglesias et al., 2013). Studies show induced inflammation in rats' brain due to manganese oxide inhalation. Silver NPs cause oxidative stress and the upregulation of oxidative stress-related genes in the cortex and hippocampus of mice (Haase et al., 2012; Feng et al., 2015).

Effect of Nanoparticles on the Endocrine System

In the past few decades, there has been a substantial rise in disorders of the immune system. One of the reasons associated with it is the exposure of workers and the general population to contaminants that exert adverse effects on endocrine-disrupting chemicals (EDCs) (e.g., CdTe quantum dots), which play a prime role in altering the hormonal and homeostatic systems (Iavicoli et al., 2013; Lu et al., 2013). A study shows how 150 μ g/kg of iron oxide NPs administered in male rats increased the T3 thyroid hormone and decreased the thyroid-stimulating hormone, thus creating a net imbalance (Jiang et al., 2019). Commencing with overstimulation, palladium NPs have shown to gradually act on hormone receptors, blocking the signal cascades (Leso et al., 2018). Some studies have observed that diabetic animals are more prone to hormonal disbalance that non-diabetic ones due to NP exposure (Li et al., 2009; Lu et al., 2013).

Effect of Nanoparticles on the Reproductive System

One of the main reasons for the recent spike in infertility cases has been attributed to the accumulation of NPs. An experimental study on high-fat diet rats shows that administering silica NPs (which are common at workplaces) decreased sperm concentration and motility rates and increased the abnormality rates of sperm (Zhang et al., 2020). TiO2 NPs exhibit genotoxic effects as they cross the blood-testis barrier and causes inflammation and cytotoxicity, thus a significant loss of sperm DNA integrity was observed (Santonastaso et al., 2019). NPs affect the reproductive system by inducing cytotoxicity in ovarian cells, thereby disrupting the process of oogenesis and eventually causing the overproduction of ROS, apoptosis, and sex-hormone imbalance in the body (Iavicoli et al., 2013). In a study, an animal administered with MoO3 showed a significant decrease in the right ovary's weight and uterine weight, which ultimately led to an adverse effect on the reproduction process (Asadi et al., 2019). Hence, NPs are significantly responsible for decreasing birth rates and causing abnormalities in offsprings. In urban areas, such cases are commonly found in females who are exposed to air pollutants, particularly particulate matter (especially NPs), due to their daily lifestyle.

RESPIRATORY MODELS

Specific models have been devised to understand the structure and effects of NPs on individual organs. The Weibel bifurcating tubes' model with 23 bifurcation units describes the branching pattern of lungs, i.e., the 23 levels of bifurcations-G0-G23. The first 16 bifurcations (G0-G16) comprise the conducting zone, while the rest (G17-G23) comprise the respiratory zone involved in the exchange of gases. This model helps in understanding the structure of the airway path (Qiao et al., 2015), and models similar to the organ have been prepared to study the effects of NPs on other body systems. To comprehend and quantify the rates and mechanism of particle deposition, clearing, and impact, the models were divided into experimental and computational methods (Qiao et al., 2015). The experimental methods include in vivo and in vitro experiments on a model organism exposed to different doses of NPs to determine deposition, clearing, and health impacts (Ji and Yu, 2012; Qiao et al., 2015).

A computational method like computational fluid dynamics (CFD) is primarily used an effective tool in predicting the airflow, particle transport, deposition, and the particle number concentration in fine particles. It is also used in the field of NP instrumentation, evolution of NP dynamics in different environment (human respiratory tract, workplaces, aerosol transport/delivery system, energy systems, etc.), and NP synthesis (Qiao et al., 2015).

Experimental Models

Respiratory tract dosimetry is a biologically based, mechanistic method that is a useful experimental technique to estimate the exposure concentrations of an inhaled substance in a model organism (Kuempel et al., 2006) that can be extrapolated to human equivalent exposure levels by adjusting the lung mass and lung surface area (Kuempel et al., 2006; Ji and Yu, 2012). The multiple-path particle dosimetry model (MPPD) developed by the Chemical Industry Institute of Toxicology (CIIT, currently known as the Hamner Institutes for Health Sciences) and the Dutch National Institute for Public Health and the Environment (RIVM) is used in the assessment of deposition and clearance of course to ultrafine particles (Ji and Yu, 2012). MPPD provides a quick, inexpensive, and efficient means to assess the internal estimates of tissue dose. This model is an efficient tool to determine the percentage change in the particulate load of respirable deposits in the human respiratory system. MPPD estimates particle deposition in the tracheobronchial region where most of the respirable particulate deposition occurs as compared with the pulmonary region (Goel et al., 2021). As the respiratory tract is divided into various regions based on anatomical function, sub-models of ventilation and aerosol transport are constructed (Asgharian et al., 2014). Settings like flow characteristics, orientation, inspiratory fraction, and parameters including multiple respiratory volumes can be altered to meet the needs of the experiment (Manigrasso et al., 2019). Another popular *in vitro* model is the air-blood barrier (ABB) models that elucidates the biological mechanisms related to the potential effects of inhaled nanoparticles. In this model, the effect of NPs on the cell lines of respiratory epithelium, endothelium, and monocyte cells are studied (Bengalli et al., 2017).

Computational Models

The CFD method is mostly used to highlight the impact of several factors including upstream flow, inlet profiles, and turbulent enhanced dispersion (Longest and Vinchurkar, 2007). Like MPPD, the airway is divided into subdivisions called tiny meshes that can help quantify the deposition of particles (Longest and Vinchurkar, 2007; Garcia and Kimbell, 2009; Qiao et al., 2015). Two approaches have been used to investigate the deposition study. First is the Eulerian approach that uses a constant laminar flow rate with non-stop fluid process, dilute particle phase (treated as interpenetrating fields), Brownian motion (depending on the concentration gradient), and a diffusion coefficient (depending on particle size). This model is used to calculate the deposition efficiency of various particle sizes. Deposition

efficiency (DE) can be calculated using the following equation:

$$DE = 1 - \left(0.819e^{-14.63\Delta} + 0.0976e^{-89.22\Delta} + 0.0325e^{228\Delta} + 0.0509e^{125.9\Delta^{\frac{2}{3}}}\right),$$

where $\Delta = \frac{DL_{pipe}}{4R^2U_{inlet}}$,

L_{pipe} is the pipe length, U_{inlet} is the inlet velocity, and R is the pipe radius. This approach neglects particle inertia and is effective for studying a large number of particles (Inthavong et al., 2011). The assumption of laminar flow is supported by the low Reynolds number (Re) (Longest and Vinchurkar, 2007; Garcia and Kimbell, 2009). The second approach is called the Lagrangian approach that considers all the above variables along with particle inertia. This is effective in studying individual particle motion. The trajectories of each particle are traced by integrating a force balance equation on the individual particles:

$$\frac{du_i^p}{dt} = F_D + F_g + F_B + F_L + F_T \tag{1}$$

Here, F_g is the gravity term; F_D is the drag force per unit particle mass; F_B is the amplitude of the Brownian force component; F_L is the lift due to shear; and F_T is the thermophoretic force. In comparison, the Eulerian model is less computationally demanding due to the single convection–diffusion equation that controls the fluid–particle interrelationship (Inthavong et al., 2011).

Apart from the aforementioned models, the World Health Organisation (WHO) recently introduced new models for the assessment of air quality. Air quality models like the AirQ model and the AirQ 2.2 software are currently in use (Conti et al., 2017). Air quality models use mathematical and numerical methods to simulate the physical and chemical processes caused by air pollutants that trigger their dispersion and chemical transformation in the atmosphere. The purpose of these models is to relate the effects of emission sources with the pollutant concentrations and to cheque whether they are crossing the standard limits or not.

CONCLUSION

The health effects of nanoparticles are well-known, and their exposure can cause serious respiratory and cardiovascular diseases. Epidemiological and toxicological studies have mentioned that nanoparticles are much toxic than coarser particles due to their ultrafine size, chemical reactivity, and longer residence time in the atmosphere. It has also be observed that as particles decrease in size, their ability to penetrate deep inside the lungs increases. The source of nanoparticles varies from anthropogenic activities (vehicular emission, industrial emission, welding fume, cigarette smoke, etc.) to natural activities (forest fire, dust storm, volcanic eruption, etc.). The transport of NP in the lungs is explained with the Weibel bifurcating tubes' model, classified into the conducting zone and respiratory zone. Owing to the ultrafine size and high retention time, nanoparticles diffuse and accumulate in the alveolar region, and some pass through the alveolar epithelium and capillary endothelial cells to enter the cardiovascular system and other internal organs. Different types of clearance mechanisms are exhibited by the human body to counter the effect of NP exposure and protect the body before the onset of the disease. Mucociliary escalation clears the upper respiratory tract, while macrophages with certain proteins help ingest the deep deposited material. Regardless, insoluble NPs get bioaccumulated in the respiratory zone over time, causing lung burden. Such NPs are attacked by phagocytes as a part of the immune response. The exposure to NPs cause the generation of ROS, resulting in cytotoxicity that leads to genotoxicity and tumourigenesis. The overproduction of ROS and the weakening of antioxidant defence system cause oxidative stress, known to trigger the release of more pro-inflammatory hormones that lead to inflammation as well as acute and chronic lung diseases. Thus, NPs cause damage in single-stranded and double-stranded DNA or deletions of its parts and induce mutations in the genetic material, generating neoplasms and carcinomas in the lungs. Besides the harm caused in the lungs, NPs also cause damage in the other body systems including the cardiovascular and gastrointestinal system and lead to cardiac arrest and ulcer, respectively. Comorbid individuals are always at a higher risk of several health complications due to NP accumulation.

This is the first comprehensive review about NPs' source, exposure, and impact on different human body systems, focusing on the respiratory system. The review provides detailed insights about the complex mechanism of NP transportation, clearance, and deposition in lungs that cause lung burden after long-term NP exposure. It offers a detailed overview of the influence of NP exposure in causing toxicological, genotoxic, and tumourigenic effects on the human body. It also mentions the experimental and computational respiratory models, which can be important tools to understand NP's transportation, clearance mechanism, and deposition patterns in human body.

AUTHOR CONTRIBUTIONS

SSo wrote the full text of manuscript. SSo and PS involved in conceptualization and design of the work. SM and JA contributed in acquisition, analysis and interpretation of data for the work, and helped in manuscript writing. SSu involved in acquisition and interpretation of data and critically reviewed the manuscript. DR, NM, and TV performed the whole formatting and editing of manuscript. PS coordinated and supervised the entire team. All authors contributed to the article and approved the submitted version.

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Effect of Lockdown Amid COVID-19 on Ambient Air Quality in 16 Indian Cities

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The COVID-19 pandemic has affected severely the economic structure and health care system, among others, of India and the rest of the world. The magnitude of its aftermath is exceptionally devastating in India, with the first case reported in January 2020, and the number has risen to \sim 31.3 million as of July 23, 2021. India imposed a complete lockdown on March 25, which severely impacted migrant population, industrial sector, tourism industry, and overall economic growth. Herein, the impacts of lockdown and unlock phases on ambient atmospheric air quality variables have been assessed across 16 major cities of India covering the north-to-south stretch of the country. In general, all assessed air pollutants showed a substantial decrease in AQI values during the lockdown compared with the reference period (2017-2019) for almost all the reported cities across India. On an average, about 30-50% reduction in AQI has been observed for PM_{2.5}, PM₁₀, and CO, and maximum reduction of 40–60% of NO₂ has been observed herein, while the data was average for northern, western, and southern India. SO2 and O3 showed an increase over a few cities as well as a decrease over the other cities. Maximum reduction (49%) in PM_{2.5} was observed over north India during the lockdown period. Furthermore, the changes in pollution levels showed a significant reduction in the first three phases of lockdown and a steady increase during subsequent phase of lockdown and unlock period. Our results show the substantial effect of lockdown on reduction in atmospheric loading of key anthropogenic pollutants due to less-to-no impact from industrial activities and vehicular emissions, and relatively clean transport of air masses from the upwind region. These results indicate that by adopting cleaner fuel technology and avoiding poor combustion activities across the urban agglomerations in India could bring down ambient levels of air pollution at least by 30%.

Keywords: lockdown, COVID-19, air quality, Indian cities, mitigation

INTRODUCTION

A novel infectious disease for the first time was identified in Wuhan, China, in late December 2019 and was named as 2019 novel corona virus and later on renamed as the COVID-19 (Chen et al., 2020) on February 11, 2020 by the International Committee on Taxonomy of Viruses. Later on in January 2020, the World Health Organization (WHO) revealed human transmission of COVID-19

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through respiratory droplets (WHO, 2020), which subsequently got spread throughout the China, and the outbreak was turned into an epidemic (Dutheil et al., 2020). On January 30, 2020, the WHO declared the COVID-19 as a global pandemic. Subsequently, the transmission of pandemic COVID-19 *via* airborne pathway was recognized by the WHO. After the severe acute respiratory syndrome coronavirus (SARS-CoV) in 2002, which influenced around 37 countries and the Middle East respiratory syndrome coronavirus (MERS-CoV) in 2012, the COVID-19 is the third major zoonotically spread calamity of the current century.

To slow down the rate of spread of virus, almost all the countries have followed partial-to-complete lockdown practice (Tosepu et al., 2020). Globally, the economic activities were ceased, and stock markets plunged along with the falling carbon emission. The industrial activities were shut down globally due to the imposed lockdown. The informal economic sector suffered a major fall along with the transport sector as most of the countries imposed complete lockdown. Global fossil fuel demand dropped down severely, as industrial and transport sectors came to a halt for a while across the world. As far as the scenario of COVID-19 in India is concerned, the first COVID-19 incidence in India was registered on January 30, 2020, in the state of Kerala, and the travel restrictions to several countries were imposed from March 11, 2020 soon after a steep rise in the number of incidences on March 4, 2020. From March 16 onward, all places of public gathering were shut down across India. The first nationwide lockdown, on a trial mode, was witnessed on March 22, 2020, and subsequently, from March 24, 2020 a nationwide complete lockdown for 21 days was announced by the Central Government of India. The different phases of lockdown and unlock in India are given in Table 1.

Significant impacts of lockdown were observed on air quality across the world (Berman and Ebisu, 2020; Nakada et al., 2020; Venter et al., 2020) and in India (Kumar, 2020; Kumar, 2020; Sarfraz et al., 2020; Sharma et al., 2020; Dumka et al., 2021). A positive association between COVID-19 cases and meteorological parameters has also been shown recently (Kumar, 2020). Dumka et al. (2021) has shown that more than 50% reduction in PM2.5 and NO2 concentrations occurred over Delhi-NCR, mainly due to restrictions in traffic-related activities. Nationwide lockdown amid the COVID-19 pandemic had a significant impact on the air quality index. Thus, a quantitative assessment of air quality variables is needed to understand the impact of lockdown on anthropogenic emission source impact and their reframe mitigation policies in India. The major objective of the present study was to assess the changes in ambient air quality during lockdown and in the subsequent unlock phases across India.

Data and Methodology

To assess the impact of nationwide lockdown on ambient air quality in India, we have chosen 16 major cities (Chandigarh, Delhi, Jaipur, Lucknow, Patna, Kolkata, Gandhinagar, Bhopal, Nashik, Mumbai, Nagpur, Hyderabad, Bengaluru, Chennai, Visakhapatnam, and Thiruvananthapuram) across the country (**Figure 1**). The selection of these cities is based on the level of urbanization linked to air pollution, implementation of lockdown policy, coverage of typical major cities in the country, and availability of data set of air pollution and meteorological parameters. The daily average (and median) along with minimum, maximum, and standard deviation of each of the air pollutants and meteorological parameters during January 1 to June 30 for 4 years (2017-2020) were retrieved from the World Air Quality Index (WAQI) Project (https://aqicn.org/data-platform/covid19/) having data source (for Indian region) originally from India's Central Pollution Control Board (cpcb.nic.in/), U.S. Embassy and Consulates' Air Quality Monitor in India (in.usembassy.gov/embassyconsulates/new-delhi/air-quality-data/), Delhi Pollution Control Committee (dpccairdata.com/), and India Meteorological Department (www.imd.gov.in/). The daily average air pollutants and meteorological data set for each of the cities have been deduced from the average of the respective data recorded from monitoring stations located in that city. The key air pollutant species such as particulate matter with aerodynamic diameter \leq 2.5 and \leq 10 μ m (PM_{2.5} and PM₁₀), nitrogen dioxide (NO₂), sulfur dioxide (SO₂), carbon monoxide (CO), and tropospheric ozone (O₃) along with the meteorological parameters [relative humidity (RH), near surface air temperature (T), and wind speed (WS)] have been assessed herein.

All air pollutants reported in this study were converted with respect to the United States Environmental Protection Agency (US EPA) standard (Mintz, 2018). The first step is to identify the highest concentration of pollutants among all of the monitors within each locations and then truncate it as PM_{2.5} (μ g/m³) to one decimal place, PM₁₀ (μ g/m³) to integer, CO (ppm) to one decimal place, SO₂ (ppb) to integer, NO₂ (ppb) to integer, and O₃ (ppm) to three decimal places. Subsequently, with the aid of the two breakpoints, the concentration was summarized (**Supplementary Table 1**). Finally, the index was calculated using the following equation:

$$I_p = \frac{I_{Hi} - I_{Lo}}{BP_{Hi} - BP_{Lo}} \left(C_p - BP_{Lo} \right) + I_{Lo} \tag{1}$$

where, I_p is the index of a pollutant p, C_p is the truncated concentration of that pollutant p, BP_{Hi} is the concentration breakpoint $\geq C_p$, BP_{Lo} is the concentration breakpoint $\leq C_p$, I_{Hi} is the AQI value corresponding to BP_{Hi} , and I_{Lo} is the AQI value corresponding to BP_{Lo} . The details of AQI estimation for different pollutants have been provided elsewhere (Bishoi et al., 2009; Mintz, 2018).

The concentration of the ambient air pollutant not only depends on the intensity of its emission but also on meteorological conditions. Therefore, in order to see the net change due to restrictions on emission sources, we need to normalize the concentration of the pollutant with meteorology. There are various methods existing to account for (or normalize) meteorological effects on the concentration of the ambient air (Dai et al., 2020; Petetin et al., 2020; Falocchi et al., 2021). In this study, we have used the ventilation coefficient (VC) to normalize the meteorological influence (Dai et al., 2020)

TABLE 1	Different timelines	that India adopted	for lockdown ar	d unlock phases.
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Phases	Timeline	Duration	Activities restricted/permitted
Phase 1	March 25, 2020 to April 14, 2020	21 days	All industries and services suspended except essential services
Phase 2	April 15, 2020 to May 3, 2020	19 days	Conditional relaxation from April 20 for the COVID-contained regions. Classification of regions into red, orange, and green zones. Agribusinesses, public work programs allowed to reopen with social distancing. Cargo transportation vehicles, including trucks, trains, and planes were started. Banks and small retail shops were reopened. Interstate movement of stranded allowed
Phase 3	May 4, 2020 to May 17, 2020	14 days	On May 1, red zones (130 districts), orange zones (284 districts), and green zones (319 districts)
Phase 4	May 18, 2020 to May 31, 2020	14 days	States control the demarcation of green, orange, and red zones. Red zones further divided into containment and buffer zones
Unlock			
Unlock 1.0	June 1, 2020 to June 30, 2020	30 days	First phase of reopening shopping malls, religious places, hotels, and restaurants. Large gatherings were still banned, no restrictions on interstate travel. Night curfews to be in effect
Unlock 2.0	July 1, 2020 to July 31, 2020	31 days	Lockdown measures were imposed in containment zones. In all other areas, most activities were permitted. Night curfews to be in effect. Limited international travel has been permitted as part of the Vande Bharat Mission



on ambient AQI. The hourly wind vector at 2 m (above the ground) and mixing-layer height data over all studied 16 cities (during March to June, 2017–2020) have been obtained from ERA5 global reanalysis products (Hersbach et al., 2020). The daily mean VC is estimated by taking the product of the daily mean mixing-layer height and wind speed for each of the cities during the study period. Furthermore, climatological or the long-term mean of VC (VC_{mean}) specific to each city has been estimated by taking the average of daily VCs during the study period (2017–2020). The influence of meteorological dispersion on observed AQI has been removed by normalizing the AQI data of each pollutant with VC_{mean}
for every city using the following equation (2) as given by Dai et al. (2020).

$$AQI_{VC,i} = AQI_i^*(VC_i/VC_{mean})$$
(2)

where $AQI_{VC,i}$ is meteorology normalized AQI for the ith day, AQI_i is the AQI measured on the ith day, and VC_i is the ventilation coefficient on the ith day. Furthermore, meteorologically normalized (hereafter represented as "met normalized") AQI data of each pollutant has been used to see the relative changes in AQI during the lockdown period compared with the reference period across the 16 Indian cities.

RESULTS AND DISCUSSION

Effect of Meteorology on Observed Changes in Ambient AQI

The wind speed and direction are important parameters that significantly impact the concentration of the pollutants in ambient air. We have analyzed the wind rose maps for reference and lockdown period for each city. Supplementary Figure 1 shows the prevailing wind direction and speeds in selected cities in north India for the study period. The suffixes "Ref" and "Cov" refer to reference period (in 2017, 2018, and 2019) and lockdown period due to COVID (in 2020) trends, respectively. The legend colors represent increasing wind speed from top to bottom. The wind rose plots for other cities are provided in the Supplementary Material for western Indian cities (Supplementary Figure 2) and south Indian cities (Supplementary Figure 3) in our study. Briefly, we have not found any significant change in prevailed wind directions during the COVID year with respect to reference period over any city. In other words, we can say that both the periods have a similar kind of wind pattern over the Indian cities. Furthermore, Table 2 shows the mean values of meteorological parameters (RH, T, and WS) during the reference and lockdown period for the 16 major cities of India. In general, the lockdown period was, on an average, characterized by relatively high humidity and lower mean temperature compared with the reference period. The wind speed appeared to be nearly similar during lockdown as well as the reference period. Nevertheless, the day-to-day variability in meteorological parameters for 1 year to another could be quite significant, and thus, taking into account for meteorological variability, while comparing pollutant concentrations is of utmost importance (Dandotiya et al., 2019; Nandi, 2020).

Table 1 shows the different phases of lockdown and unlock period practiced in India to control the spread of COVID-19. Lockdown period was imposed in four phases: P1 (March 25 to April 14), P2 (April 15 to May 3), P3 (May 4 to May 17), and P4 (May 18 to May 31) (Saha and Chouhan, 2021). P1 and P2 were the periods of complete nationwide lockdown, whereas P3 and P4 were the periods of partial lockdown. In order to assess the maximum reduction in anthropogenic contribution to AQI, we choose the complete lockdown period. The mean USEPA standardized AQI values (with and without normalization to meteorological parameter) of various air pollutants averaged

TABLE 2 | Mean values of meteorological parameters during complete lockdown period (in 2020, as Cov.) and reference period (as Ref.).

	RH	(%)	Т ((° C)	WS (m/s)		
	Ref.	Cov.	Ref.	Cov.	Ref.	Cov.	
Chandigarh	45.3	55.3	28.1	23.4	1.5	1.6	
Jaipur	21.4	32.5	31.9	29.2	1.3	1.1	
Delhi	34.6	47.6	29.8	27.8	1.6	1.4	
Lucknow	39.1	47.3	31.5	29.6	0.5	0.6	
Patna	53.3	56.7	31.6	27.6	0.8	0.6	
Kolkata	71	68.7	28.3	28.1	1.4	0.7	
Gandhinagar	28.4	40.3	33	30.5	1.9	1.1	
Nashik	33.4	38.6	27	27.9	2.5	2.2	
Nagpur	-	49.2	-	30.7	-	0.4	
Mumbai	65.6	76.1	30.3	30	0.9	1.3	
Bengaluru	44.5	47.4	26.9	25.7	1.6	1	
Hyderabad	43.9	47.2	29.4	30	1.3	1	
Chennai	64.3	71.1	30.6	29.7	-	2.6	
Bhopal	23.4	31.3	29.7	30.9	0.7	0.8	
Thiruvananthapuram	71.9	76.6	29.4	28.3	1.5	1	
Visakhapatnam	72.6	71.9	28.4	32.2	2.9	2.2	

during March 25 to May 3 for the reference and lockdown period for the 16 major cities of India are shown in Table 3. Figure 2 shows city-specific percent change of key pollutants (with and without normalization to meteorological parameter viz. VC) averaged over complete lockdown period (phase 1 and phase 2: March 25 to May 3, 2020) compared with the reference timeperiod for the 16 major cities across India. Table 3 shows that the AQI values of key air pollutants get substantially increased after met normalization during the reference period over almost all Indian cities, whereas minimal effect has been observed during the lockdown period. Moreover, percent changes in key pollutants averaged over the complete lockdown period (Figure 2) show higher reduction for met normalized data compared with that of without met normalization. If we take only without met normalized data, on an average, more than 30% decrease has been observed in PM2.5 (-35% averaged over all cities), PM_{10} (-37% averaged over all cities), and CO (-32% averaged over all cities) over most of the cities in India. The maximum negative change is found in NO₂ AQI over all the cities with about -42% averaged over India. From Figure 2 and Table 3, it is obvious that the average difference in values for with and without met normalization of air pollutants is $11 \pm 3\%$. Furthermore, in general, with met normalized values were lower than without met normalized values. In the further discussion, we would be utilizing with met normalized values of AQI of air pollutants.

Effect of Lockdown on Citywise Ambient AQI

In general, all the air pollutants (met normalized) showed a substantial decrease in AQI values during COVID period compared with the reference period for almost all the assessed cities across India. On an average (over all cities), more than 24% TABLE 3 | Mean EPA AQI values of various air pollutants (with and without normalization to meteorological parameter) during complete lockdown period (in 2020, as Cov.) and reference period (as Ref.).

		PI	M _{2.5}	P	M ₁₀	s	02	c	0	N	1 O 2	(O ₃
		Ref.	COV.	Ref.	COV.	Ref.	COV.	Ref.	COV.	Ref.	COV.	Ref.	COV.
Chandigarh	Without met	112	72	_	46	3.0	5.8	4.5	3.1	9.3	6.5	16.8	12.5
	With met	158	68	-	43	4.0	5.5	6.0	2.8	13.4	6.0	20.2	12.0
Jaipur	Without met	134	77	88	51	4.5	5.5	5.9	4.9	12.8	5.6	20.7	21.7
	With met	184	72	119	49	5.7	5.0	7.5	4.4	16.9	5.2	25.2	18.9
Delhi	Without met	163	106	142	68	7.9	6.0	7.9	6.0	19.0	7.2	13.9	15.2
	With met	240	106	211	69	11.7	6.1	11.3	6.1	27.9	7.3	21.7	15.7
Lucknow	Without met	97	76	68	53	11.2	2.9	12.7	3.4	11.4	2.8	10.3	8.9
	With met	127	89	89	62	14.6	3.5	16.4	4.1	14.8	3.3	13.0	11.0
Patna	Without met	158	98	-	61	7.0	4.7	8.9	6.1	8.0	7.4	13.4	15.9
	With met	181	113	-	73	7.9	5.6	10.0	6.9	9.2	8.5	14.8	16.5
Kolkata	Without met	114	80	62	42	2.9	4.1	4.9	3.3	10.3	5.0	10.8	17.0
	With met	165	83	89	45	4.1	4.3	7.4	3.2	14.9	4.9	14.5	17.5
Bhopal	Without met	133	87	118	53	9.6	5.2	10.3	4.6	15.0	4.6	15.3	10.4
	With met	156	89	143	55	10.9	5.2	11.6	4.7	17.0	4.6	17.5	10.0
Gandhinagar	Without met	165	81	-	57	25.3	3.6	7.2	5.7	28.1	5.5	11.9	6.4
	With met	160	77	-	54	25.7	3.5	6.9	5.5	27.9	5.0	12.3	6.3
Nashik	Without met	121	84	65	45	2.5	1.2	4.6	3.5	10.7	9.5	33.6	25.4
	With met	132	76	72	42	2.7	1.2	4.9	3.3	11.3	8.9	35.8	22.2
Nagpur	Without met	135	71	64	38	6.0	0.8	8.0	3.6	11.7	8.5	26.3	18.4
	With met	208	70	99	37	8.3	0.7	11.7	3.6	17.0	8.2	43.4	19.0
Mumbai	Without met	171	122	-	-	3.8	2.7	9.1	9.1	16.3	6.2	21.8	10.3
	With met	153	119	-	-	3.4	2.6	8.1	8.9	13.9	6.1	19.8	9.8
Bengaluru	Without met	111	75	64	43	2.2	3.3	6.2	6.1	10.9	5.1	17.3	16.5
	With met	115	81	68	47	2.4	3.4	6.2	6.3	11.0	5.1	18.7	17.3
Hyderabad	Without met	125	89	80	50	4.3	2.2	6.0	3.8	13.8	6.4	21.0	11.1
	With met	140	87	90	49	4.6	2.1	6.6	3.6	15.0	6.3	22.6	10.8
Visakhapatnam	Without met	98	57	67	44	4.7	3.3	7.1	4.1	11.4	11.5	5.2	7.7
	With met	122	56	82	44	6.3	3.3	9.0	3.9	13.9	11.3	6.1	7.6
Chennai	Without met	87	49	-	8	2.7	2.2	7.7	5.6	6.3	2.6	9.6	10.3
	With met	96	43	-	7	3.0	2.0	8.5	5.4	7.2	2.2	10.9	10.1
Thiruvananthapuram	Without met	85	67	50	39	2.1	2.6	8.1	4.8	3.6	3.5	17.6	14.0
	With met	92	62	56	36	2.1	2.5	8.1	4.5	3.5	3.5	18.7	13.7

decrease has been observed for all the AQI of the pollutant; PM2.5 decreased by 45%, PM₁₀ decreased by 48%, and CO decreased by 41%. The maximum decrease has been found for NO₂ AQI over all the cities with an overall decrease of 52% when averaged for all 16 cities. The decrease in PM pollution and gaseous pollutants (CO and NO₂) clearly reflects the impact of ceased industrial and vehicular activities during lockdown. SO2 and O3 were also decreased in the lockdown period compared with the reference period, except over a few cities wherein these species showed an increase in their concentration during the lockdown period with respect to the reference period. For example, the SO₂ AQI showed a statistically significant increase in Chandigarh (34.4%, from 4.0 \pm 2.0 to 5.5 \pm 3.2; two-tailed *t*-value: 3.5), Bengaluru (45%, from 2.4 \pm 1.0 to 3.4 \pm 1.5; two-tailed *t*-value: 4.8), and Thiruvanthapuram (15.4%, from 2.1 ± 0.8 to 2.5 ± 1.0 ; two-tailed *t*-value: 2.6) in the COVID period compared with the reference period. Mor et al. (2021) has also reported an increase of 2-20% in SO₂ concentration during different lockdown phases compared with the pre-lockdown period over Chandigarh. The increased concentration was attributed to atmospheric transport of SO₂ emissions from coal-based thermal power plants upwind of the measurement location (Mor et al., 2021).

To investigate further about the increase in SO_2 during COVID lockdown in our study over these three cities, we have analyzed the bivariate polar plots for the reference period and COVID period (**Figure 3**). Polar plots suggest that neither the wind speed nor the wind direction was significantly different at Chandigarh, Bengaluru, and Thiruvanthapuram during the COVID year with respect to the reference period. However, the AQI of SO₂ was substantially higher during the lockdown period amid COVID-19 compared with the reference period (**Figure 3**). Though we could not find anywhere the energy



demand estimations for these cities during the lockdown period, we have tried to collect the supporting information for the observed increase in SO₂ levels during the lockdown period. In the wake of the COVID-19 pandemic, the Indian government, in collaboration with established textile industries, started domestic manufacturing of personal protective equipment (PPE) in India since March 2020 (Lakshmanan and Nayyar, 2020). Thermal energy consumption in textile manufacturing units is about 70– 80% in India (Bhaskar et al., 2013). Moreover, these coal-based thermal power sources in textile industries is the major source of SO₂ in the atmosphere (Rabbi, 2018; Niinimäki et al., 2020).

Since all other industries were shut down during the lockdown period, we therefore are attributing the textile industries, involved in PPE kit manufacturing, as the major energy consumption units in a particular city during the lockdown period. The major hubs identified for PPE kit manufacturing were Ludhiana, Bengaluru, and Ernakulum, among others (Kitex Garments ltd., 2020; The News Minute., 2020; The Tribune., 2021). Thus, it is logical to state here that there would have been higher energy consumption in

Ludhiana, Bengaluru, and Ernakulum. The three cities in which we observed increased SO₂ levels during lockdown were Chandigarh, Bengaluru, and Thiruvananthapuram. Ludhiana and Ernakulum are situated in the north-west direction of Chandigarh and Thiruvananthapuram, respectively. Thus, prevailed north-westerly winds would have favored the transport of SO₂ to Chandigarh from power plants feeding the energy to textile industries in Ludhiana (Figure 3). Likewise, prevailed north-westerly winds would have favored the transport of SO₂ to Thiruvananthapuram city from power plants feeding the energy to textile industries in Ernakulum. However, Bengaluru itself was one of the major hubs for the manufacturing of PPE kits in south India (The News Minute., 2020). This would have led to higher energy consumption in the city, which would have led to higher emissions of SO₂ from nearby power plants, the effect of which was observed in our results.

We have also witnessed a significant increase in O_3 over Kolkata (~18%, from 14.5 \pm 4.5 to 17.5 \pm 11.6; two-tailed *t*-value: 2.4) and Visakhapatnam (~20%, from 6.1 \pm 2.2 to 7.6 \pm 4.2; two-tailed *t*-value: 2.9) during the lockdown period



compared with the reference period. Dumka et al. (2021) has shown about 4–8% increase in O₃ concentration over Delhi— NCR, mainly related to NOx chemistry. Pathakoti et al. (2020) has shown a dip of ~17–18% in mean NO₂ levels (with maximum reduction over Delhi ~54%) using satellite remote-sensing data over India during the lockdown period. Jain et al. (2021) has also shown a significant reduction (~50%) in short-lived gaseous air pollutants such as NO₂ and SO₂, whereas minimal reduction (~10%) in CO and O₃ compared with that of 2019, which has direct as well as indirect impacts of anthropogenic emissions. Venter et al. (2020) has also shown an increase in satellite-based O₃ concentrations over polluted regions of China and India and a decrease in other parts of the world during the lockdown period compared with 2019. These results may be understood as non-linear chemical interactions of volatile organic compounds (VOCs) and oxides of nitrogen (NO_x = NO + NO₂) in formation/destruction of tropospheric O₃ under different environmental conditions (Sillman, 1999). Venter et al. (2020) has suggested that the VOC limited region may experience an increase in O₃, whereas the NO_x limited region experiences a decrease in O₃. Thus, our findings on change in air pollution magnitude are consistent with the previous literature.

Effect of Lockdown on Regionwise (North, West, and South India) Ambient AQI

The aforementioned analysis indicates a significant impact of lockdown amid COVID-19 on improvement in ambient air quality in Indian cities, i.e., huge reduction in levels of air





pollutants. These reductions in AQIs were manifestation of both the weak source intensity, atmospheric chemistry, and favorable meteorological condition. Similar findings for selected air pollutants have been reported for different cities of India and from different geographical locations across the globe (Kumar et al., 2020; Mahato et al., 2020; Otmani et al., 2020; Venter et al., 2020; Dumka et al., 2021).

Since most of the cities are showing near similar trends of change in levels of air pollutants, it would be reasonable to group them regionwise and assess the improvement in air quality due to lockdown amid COVID-19 on a regional basis. We have grouped different cities into three broad regions, viz., north, western, and south India. North India was represented by Chandigarh, Jaipur, Delhi, Lucknow, Patna, and Kolkata, whereas western India was represented herein by Nashik, Gandhinagar, Mumbai, Nagpur, and Bhopal in our study. Hyderabad, Bengaluru, Chennai, Visakhapatnam, and Thiruvananthapuram were grouped as south India. Subsequently, we have averaged the data set of respective cities on a regional basis (north, west, and south) to assess the regionwise change in levels of air pollutants during pre-lockdown (Pre-Lock), lockdown (LockP1, LockP2, LockP3, and LockP4), and post-lockdown (Unlock 1) period over India. **Figure 4** shows the percent change of different air pollutants during different phases [Pre-Lock, Lockdown (P1, P2, P3, and P4) and Unlock 1] over (a) north, (b) west, and (c) south India. It shows that by-and-large, the concentrations of all pollutants were decreased (except for some species during P4) over different regions of India in the lockdown period compared with the reference period. The reduced levels of air pollutants during pre-lockdown in India may be explained on the basis of worldwide lockdown leading to reduced long-range transport or transboundary impact of air pollutants (Venter et al., 2020) as many European and Asian countries had complete to partial lockdown during January–March 2020.

The temporal variability in percentage change of different pollutants shows maximum reduction during lockdown phases over all three regions. However, different interphase (lockdown phases P1, P2, P3, and P4) trends were observed across different regions. In general, maximum reductions (for almost all pollutants) were observed in the first three phases of lockdown (P1, P2, and P3) over west and south India. The increase in residential mobility during lockdown compared with that in prelockdown is reported, which steadily decreases from P1 to P4 (~31 to 18%) over India (Saha and Chouhan, 2021). North India, having the highest air pollution in normal days among regions assessed herein, shows a quite unique pattern where we see large reductions during P2 and P3, and thereafter buildup of pollution in P4 and a drastic decrease during unlock 1 period. This result can be explained on the basis of large temporal variability in the concentration of the pollutant during a short period of P4, as evident by larger error bars in Figure 4A. Thus, our analysis shows that the first three phases of lockdown were considerably associated with decreased levels of air pollutants and further relax in lockdown starts building up of pollution over all three Indian cities. In other words, it indicates a quick replenishment of air pollution soon after the lift of lockdown. This is likely due to the increase in economic activities and transportation across the country.

Overall, maximum reduction in SO2 was observed over west India (~61%) and in NO2 (~59%) over north India, and minimum reduction in almost all pollutants has been observed over south India during the lockdown period. Higher reductions in certain species during the lockdown period amid COVID-19 could be attributed to the shutdown of its major polluting source. For example, the maximum reduction (49%) of PM_{2.5} in this study has been observed over north India. Pathakoti et al. (2020) has also observed maximum reduction in aerosol levels over the IGP region with an average reduction of \sim 24% over India. A similar kind of spatiotemporal variability in aerosol optical depth has been reported over India using satellite measurements (Soni, 2021). Nigam et al. (2021) has also found an increase in air pollution over western industrial cities of India, while restrictions were relaxed in P4 and unlock period.

CONCLUSIONS

In this study, we have made an attempt to assess the effect of lockdown amid COVID-19 on the improvement in ambient air quality in the year 2020 over 16 Indian cities. Thus how, this study covers a wide spatial coverage in ambient air quality assessment from north to south in the Indian subcontinent. In general, all the air pollutants assessed herein exhibit a remarkable decline in their abundance during the lockdown period compared with their concentrations during the previous period in the years 2017-2019 (termed as reference period). Averaging over different regions of India, the air quality showed about 30-50% reduction for PM2.5, PM10, and CO, and a maximum reduction of 40-60% for the NO₂ with significant spatial variability. The concentrations of SO₂ and O₃ exhibited both the decline as well as rise in their abundance pattern during the lockdown period compared with the reference period plausibly highlighting the role of atmospheric chemistry in regulating these reactive chemical species in the urban airshed under ambient atmospheric condition. Furthermore, we have also assessed the effect of ambient meteorology on observed reduction in air pollution using meteorologically normalized AQI values. Around 10% further reduction during lockdown period compared with the reference period was contributed only due to meteorological condition. The spatiotemporal variability of different air pollutants across India emphasizes the existence of different strengths and/or nature of emission sources in north, west, and south India. Maximum fine particulate matter reduction was observed over north India during the lockdown period. The buildup of air pollution was observed in the later phase of lockdown and unlock due to relaxed restrictions on anthropogenic activities (industrial and transport). This study urges that by adopting cleaner fuel technology and avoiding poor combustion activities (e.g., crude open biomass burning) in the urban agglomerations and rural areas within India, the ambient air pollution could be reduced by around 30-60% compared with business-as-usual levels.

DATA AVAILABILITY STATEMENT

Publicly available datasets were analyzed in this study. This data can be found here: https://aqicn.org/data-platform/covid19/.

AUTHOR CONTRIBUTIONS

AM, AS, and CS collected the required data. Data analysis and interpretation is done by AM and PR. AM and PR drafted the manuscript and final editing is done by all authors. All authors contributed to the article and approved the submitted version.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/frsc.2021. 705051/full#supplementary-material

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Nature-Based Solutions for Co-mitigation of Air Pollution and Urban Heat in Indian Cities

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The urban population is subjected to multiple exposures of air pollution and heat stress and bear severe impacts on their health and well-being in terms of premature deaths and morbidity. India tops the list of countries with the highest air pollution exposure and hosts some of the most polluted cities in the world. Similarly, Indian cities are highly vulnerable to extreme heat with the frequency of heatwaves expected to increase several-fold in urban areas in India. It is reported that mitigating air pollution could reduce the rural-urban difference of the incoming radiation thus resulting in mitigation of the urban heat island effect. Since the interaction between urban heat and air pollution is dynamic and complex, both these factors should be considered by the urban authorities in designing mitigation strategies. Given the multi-functional nature and cost-effectiveness of Nature-Based Solutions (NbS), they appear to be the most appropriate remedy for environmental issues of urban areas, particularly in developing countries. In addition to improving public health (through the reduction in air pollution and urban heat), NbS also provides a wide range of co-benefits such as reducing energy cost and health costs as well as conservation of biodiversity. This review is an attempt to understand the potentials of NbS in co-mitigating air pollution and urban heat in Indian cities. A framework for the planning and design of NbS in Indian cities is also proposed based on the review that could help city planners and decision-makers in addressing these two issues in an integrated manner.

Keywords: nature based solution, Urban Heat Island, air pollution mitigation, air pollution tolerance index, co-mitigation

INTRODUCTION

As a rapidly urbanizing nation, India faces the challenge of developing its cities sustainably. Most cities in India have been growing organically lacking prudent planning that often results in degradation of their environment and ultimately resulting in various health implications for the citizens. Urban planners and policymakers hence need to anticipate the various environmental impacts of their plans and policies to ensure sustainable urbanization. The two major issues that cities across the globe facing are: Air Pollution (Manisalidis et al., 2020) and the Urban Heat Island (Ulpiani, 2021). Interestingly, the two phenomena are also intricately linked to each other through positive feedback loops (**Figure 1**). The change in urban microclimate can affect the pollutant dispersion, thus the air quality. The Short-Lived Climate Pollutants (SLCP) such as ozone and black carbon can trap heat thus increasing the temperature. Both air quality and urban microclimate are also affected by other urban characteristics such as urban morphology and

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green cover. Hence, it is important to study the two issues together and understand their integrated exposure.

Rapid urbanization has resulted in significant changes in land use and land cover which is affecting the environment in myriad ways. Specific qualities of urban materials (e.g., low albedo and high volumetric heat capacity of concrete asphalt) (Mohajerani et al., 2017), urban morphology and design (decreased sky view factor due to densely places tall buildings) (Dirksen et al., 2019), along with the heat generated from energy usage and other anthropogenic sources (Singh et al., 2020b) increases the temperature in an urban area in comparison to the surrounding suburban and/or rural areas. The phenomenon is termed as Urban Heat Island or UHI (Oke, 1973). This works as positive feedback for escalating energy consumption through increased demands for cooling. UHI also tends to intensify the effect and impact of extreme heat and heatwave events in cities (Rizvi et al., 2019). Urbanization impacts are not only limited to direct changes in land surface and air properties, but are also characterized by increase in vehicular and industrial activities that cause further deterioration in air quality (Manisalidis et al., 2020). Rapid urbanization and industrialization also result in increased anthropogenic activities which is the main cause for the deterioration of air quality in many cities. Ample studies on changes in Land Use Land Cover (LULC) and urbanization, and its effect on urban temperature and air pollution are available in India. However, there is a significant research gap on understanding the combined exposure and mitigation of urban heat and air pollution in urban areas. This is in spite of the fact that India has a high potential for co-benefits of mitigation of air pollution and climate change.

Considering the degradative nature of the current urbanization and growth and development practices, governments across the world are increasingly investing in ecosystem-based approaches of development. Nature-based Solutions (NbS) for city planning are hence, becoming popular among policy makers and practitioners. NbS has found great applications in water and energy security, disaster management and risk reduction as well as improving social well-being in urban areas. NbS aims to produce more resilient cities through restoring nature which can support conventionally built infrastructure systems (Bush and Doyon, 2019). Given the multi-functional nature and cost-effectiveness of NbS, they appear to be the most appropriate remedy for environmental issues of urban areas, particularly in developing countries like India. However, not enough steps have been taken toward integrating NbS into the city planning process. This review is an attempt to understand the potentials of NbS in co-mitigating air pollution and urban heat in Indian cities. Based on this review, a framework for the successful implementation of NbS in Indian cities is also proposed. Such a framework could facilitate effective and efficient city planning and decision-making while addressing the two issues (discussed) in an integrated manner.

METHODOLOGY

An electronic review was carried out for peer-reviewed research articles using a combination of selected keywords ("nature-based solutions," "nature-based solution + urban," "nature-based solutions + urban heat island," "nature-based solution + air pollution," "nature-based solution + air pollution + urban heat

NbS for Co-mitigation

island"). The articles published in the last 10 years (2010–2020) were retrieved from bibliographical databases (Google scholar, Sciencedirect, Pubmed) and from the reference lists of selected articles (**Table 1**). The articles that did not focus on urban areas or cities and non-English articles were excluded. The two authors independently went through the list of abstracts and finalized the articles for review.

URBAN HEAT ISLAND EXPOSURE IN URBAN AREAS

Urban Heat Island (UHI) is a phenomenon globally experienced by urban areas, wherein the urban built-up areas exhibit higher temperatures as compared to surrounding non-urban or rural landscapes (Oke, 1973). The phenomenon has been extensively documented in various cities across the world including in China (Li et al., 2018), US (Ramamurthy and Sangobanwo, 2016), Canada (Gaur et al., 2018), Australia (Santamouris et al., 2017), countries of Europe (Arnds et al., 2017; De Ridder et al., 2017), Turkey (Dihkan et al., 2018), Iran (Haashemi et al., 2016; Weng et al., 2019), Sri Lanka (Ranagalage et al., 2018), Malaysia (Qaid et al., 2016), Philippines (Estoque and Murayama, 2017), and India (Mathew et al., 2018) to mention a few with varying socio-economic and geoclimatic factors.

UHI is studied as surface UHI (SUHI) and as atmospheric UHI (AUHI). The intensity of SUHI is often measured using remotely sensed Land Surface Temperature (LST) often derived from Landsat thermal bands (Sagris and Sepp, 2017), MODIS LST products (Sidiqui et al., 2016) and ASTER thermal data (dos Santos et al., 2017). For measuring atmospheric UHI, air temperature observations are used, which are either mobile observations recorded with a moving vehicle mounted with data logger (dos Santos et al., 2017) or the meteorological station observations (Arnds et al., 2017). UHI varies both in space and time, as a function of various factors including canyon radiative geometry, thermal properties of materials, anthropogenic heat, urban greenhouse, reduction of evaporating surfaces, reduced turbulent transfer, climate, topography, physical layout of the built environment as well as short-term weather conditions (Golden and Kaloush, 2006; Shahmohamadi et al., 2011; Santamouris et al., 2019). Understanding of these factors is crucial for designing and implementing UHI mitigation solutions.

Understanding the UHI phenomenon for a city becomes crucial as it both directly and indirectly impacts human health (Vargo et al., 2016; Heaviside et al., 2017) through increased heat stress, and inflates the city's energy demands for cooling (Lowe, 2016; Liao et al., 2017). UHI and heat stress increase the building energy consumption by increasing the cooling demand in urban spaces. Literature shows that UHI causes median increase of 19% in cooling energy consumption (Li et al., 2019). However, there are huge inter-city variations ranging from 10–120% increases. Estimating this increase becomes essential given that India is a tropical country. Kumari et al. (2021) observed that UHI formation resulted in an increase of 2,600 GWh (i.e., 11.4%) in TABLE 1 | Search results on the keywords from various databases.

Keywords	Google scholar	Sciencedirect	PubMed
"nature based solutions"	11,100	960	41
"nature based solutions" + "urban"	7,830	727	16
"nature based solutions" + "urban heat island"	1,270	159	3
"nature based solution" + "air pollution"	408	161	0
"nature based solution" + "urban heat island" + "air pollution"	190	76	0

annual average electricity consumption in eight districts of Delhi between April 2012 to March 2017.

UHI has also been documented to increase the city's exposure to heatwaves under the global climate change scenario (Founda and Santamouris, 2017). With the changing climate scenario, many countries including India are becoming prone to more intense and frequent heat waves. Cities in particular due to the phenomenon of UHI, exhibit the synergies between urban heat and heatwave, making the urban areas more vulnerable to extreme heat. According to Sharma et al. (2019), frequency of heat waves for urban areas in Delhi is expected to increase from 0.8 times each summer season in current time frame to 2.1 and 5.1 times in short- and long-term projections. In the past few years researchers have focused on understanding the change in land surface temperature (LST) and factors influencing urban areas such as in Delhi (Mohan et al., 2012; Yadav and Sharma, 2018), Ahmedabad (Mathew et al., 2018), Bengaluru (Sussman et al., 2019), Jaipur (Mathew et al., 2018), Chennai (Swamy et al., 2017), Kochi (Thomas et al., 2014), Lucknow (Singh et al., 2017) and Mumbai (Dwivedi and Khire, 2018; Dwivedi et al., 2019).

Various measures for UHI have been explored particularly with contribution from studies by Mat Santamouris (Yang and Santamouris, 2018). These include, balancing the Albedo Effect in urban area and reduction of concrete urban surface (Icaza et al., 2017), planning and development of green belt and increasing of green cover in urban area (Huang et al., 2008), and installing green and white roofs (Detommaso et al., 2020).

AIR POLLUTION EXPOSURE IN URBAN AREAS

The megacities have emerged as engines of economic growth over the years but also as highly polluted urban sheds. The global urban population is expected to grow by 1.5 % per year between 2025 and 2030. Some of the highest levels of outdoor air pollution in the world are recorded in Asian cities belonging to China and India (World Health Organization (WHO), 2016). Globally air pollution is a leading environmental risk factor resulting in an estimated 5 million deaths per year (Balakrishnan et al., 2019). According to WHO, about 80% of

the urban population worldwide is exposed to air pollutant concentrations above the prescribed limits. Air pollution and related health impacts are critical in the urban environment due to its high population density and heterogeneity in emission sources resulting in pockets of very high pollutant concentration called as hotspots. The unplanned development in cities has led to the spatial variation in emission intensity which creates these local air quality control regions or hotspots. Globally, it is estimated that around 10% of the urban population lives in such hotspots (Akhtar and Palagiano, 2017). Urban air quality is largely affected by the micro-meteorology that governs the pollutant transport and resulting in hotspots. Due to the high spatial and temporal variability, the determination of air pollution hotspots is always a challenge in urban areas. Researchers have employed monitored data (fixed monitors, mobile monitors and sensors) (Kumar et al., 2015; Menon and Nagendra, 2018), satellite data (Chitranshi et al., 2015; Mahapatra et al., 2019), modeling and combined approaches (Xie et al., 2017; Mandal et al., 2020) to identify air pollution hotpots within the cities.

Other than the air pollutant emission sources, factors like UHI and climate change also affect urban air quality. Since climate change can affect the factors that govern pollutant transport like changes in chemical reaction rates and boundary layer mixing, it can have a significant effect on the regional air quality (Ebi and McGregor, 2008). According to Intergovernmental Panel on Climate Change (IPCC), elevated temperatures can have various other effects and can contribute to air pollution and related health effects (IPCC, 2013). An increase in air pollution exposure has been linked with a reduction in neighborhood greenery in the urban areas and its association with various health impacts such as cardiovascular diseases, premature mortality and children's health has been widely reported recently (Dadvand et al., 2015; Yitshak-Sade et al., 2017; Crouse et al., 2019).

NATURE-BASED SOLUTIONS (NbS)

NbS are gaining popularity in recent years in tackling environmental issues especially climate change and air pollution due to its potential co-benefits. The European Commission has defined NbS as "Solutions that are inspired and supported by nature, which are cost-effective, simultaneously provide environmental, social and economic benefits and help build resilience" (European Commission, n.d.). NbS are sustainable options to mitigate the harmful effects of climate change and pollution, improving the health and well-being of city residents at the same time benefiting biodiversity in the most resourceefficient way. Raymond et al. (2017) has developed a framework to assess the co-benefits of NbS emphasizing the need for a cross-sectoral approach to environmental policy and planning.

NbS for Urban Heat Mitigation and Adaptation

Vegetation cover has a larger effect in controlling temperature especially in urban areas as reported in various studies (Chen et al., 2020). Improving the vegetation cover/greenery is

suggested as a way to mitigate UHI and air pollution as well as improving thermal comfort in urban areas. Native plant species tolerant to pollution with higher cooling potential are ideal choices as a common solution for the problem of air pollution and UHI. Thus, planners not only need to focus on the area under green cover, the spatial distribution of green cover but also the plant species that need to be planted when working on the mitigative solutions for air pollution and urban heat.

Green spaces have both local and global adaptation as well as mitigation impacts for climate change. They help in reducing the UHI effect at the city scale as well as reinforce carbon sequestration contributing to the mitigation of global climate change. These are often projected as a "soft engineering" strategy for climate adaptation (Kitha and Lyth, 2011) and also help in climate change adaptation and disaster risk reduction by providing ecosystem services (Munang et al., 2013). Such ecosystem-based approach is often widely reported as a costeffective tool in climate adaptation.

In addition, green cover and spaces offer numerous environmental advantages. They help improve the hydrology by preventing surface runoff (Zhang et al., 2015), as well as providing ground water recharge (Ramaiah and Avtar, 2019). These could also act as buffer to extreme events such as floods and help in providing climate adaptation by acting as natural storm water drains, thus reducing climate related disaster risks for cities (Bai et al., 2018). In terms of morphology, large green spaces areas with single composition have greater cooling effects (Kong et al., 2014). Various landscape matrices have been explored to understand the significance of morphology of the urban greens. LSI or the Landscape Shape Index is an important defining factor of the morphology of a green area in the city. For small sized green spaces with complex shapes, cooling intensity is usually limited or even negative in some instances (Jaganmohan et al., 2016). Apart from size, the shape of the green areas also influences their capacity to cool. For instance, circular green space has been observed to dissipate more heat than the square one (Yu et al., 2018). In addition, the vegetation configuration is also observed to influence the cooling capacity of the green spaces. Green spaces with trees have stronger cooling effect than grass. Moreover, in London, cooling distance has been found to positively influenced by the canopy height (Monteiro et al., 2016).

NbS for Improving Air Quality

Nature-based approaches could be effectively adopted as a cheaper and sustainable option in reducing exposure to particle and gaseous pollution in urban areas. can be used for monitoring air quality as well as mitigating air pollution. Some of the sensitive species such as lichens, algae, and trees have been used as bio-indicators of air quality. At the same time, some of the plant species have the potential to reduce air pollution through mechanisms such as bioaccumulation and deposition. The tolerance and sensitivity of the plant species vary depending on the type of stress (pollutant) and plant physiology. Air pollution Tolerance Index (APTI) is a measure developed by Singh and Rao to access the plant's tolerance to air pollutants (Singh and Rao, 1983). The plant species with high APTI

value can be used to mitigate air pollution and the species with low APTI value can be used as bio-monitors due to their high sensitivity. Other than APTI, several other factors need to be considered depending on the requirement such as canopy structure, type of habitat, economic value etc. The trees with a good canopy and laminar structure can capture more dust and would be useful in removing dust. This is especially important in dusty environments such as roadsides. Tree species such as Dalbergia Sissoo and Polyalthia longifolia have good canopy structure as well as moderate to high APTI value. Meanwhile, tree species such as Mangifera indica and Azadirachta indica have economic value as well. Some of the studies have explored the possibility of estimating a new index taking into account all these factors which would help in identifying the ideal species (Pathak et al., 2011; Pandey et al., 2015a). The potential of some of the plant species in accumulating pollutants such as heavy metals has also been studied (Kumar et al., 2021). The metal accumulation species can be used in the green belt development in highly polluted areas such as industrial areas. Hence the careful selection of plant species based on their tolerance, physiological characteristics, and habitat will help in the mitigation of air pollution to a greater extent (Barwise and Kumar, 2020). Some of the commonly studied tree species for their air pollution tolerance and sensitivity in urban and industrial areas are given in Table 2.

In recent years, the term Green Infrastructure (GI) has been largely used by researchers referring to the introduction of vegetation into the urban landscape in the form of parks, green roofs, and walls and its effectiveness in reducing air pollution through various processes such as dispersion and deposition and climate change mitigation and adaptation. The World Health Organization(WHO) has recommended a green space per capita of 9 sq.m per person to improve the citizens' quality of life. Several countries such as UK and Malyasia have adopted standard-based approaches in ensuring green space availability. However, India lacks any stringent norms or standard-setting the minimum required green space per capita in urban areas. The urban Greening Guidelines 2014 by the Ministry of Urban Development suggested steps for the protection of trees and integrating green spaces in urban planning and development phases. Rapid urbanization has resulted in a considerable reduction of green cover in major Indian cities over the years. While some of the cities like Delhi have taken significant measures in restoring green spaces, most of the other cities still show a decreasing trend in their green cover. The green spaces in urban areas can be planned either by (a) using/restoring existing green areas or (b) designing new areas to meet the specific purpose in the form of parks, gardens, urban forests, roadside avenues, or vertical greening systems and green roofs.

Vertical Greening Systems (VGS)

VGS are structures where the vegetation is allowed to grow and spread over a wall or building. The potential of VGS in reducing the temperature, improving thermal comfort, energy savings and air pollution mitigation has been studied widely in the recent past (Pérez et al., 2014; Pandey et al., 2015b; Bustami et al., 2018). VGS such as green facades and green walls can reduce the temperature by passive cooling and has been identified as a solution for urban heat islands. VGS will further help in energy savings by providing thermal insulation through its shade effect (reducing solar radiation), cooling effect (evapotranspiration of plants) and insulation effect (due to insulation of different layers) (Price et al., 2015). Based on the type of plants selected, these systems can mitigate air pollution as well. Some of the climber species with high APTI have been identified as ideal plant species for the development of VGS for air pollution mitigation (Pandey et al., 2015b). Thus, if carefully selected and designed, VGS can provide solutions to both climate change and air pollution in urban areas. Vertical gardens or VGS have recently gained much attention with most of the Indian cities installing them in highly polluted areas such as roadsides.

Green Roofing

Green roofing refers to the vegetation grown over the roof of a building. Green roofs can mitigate air pollution, UHI and noise, reduce energy consumption and, manage runoff water (Vijayaraghavan, 2016; Abhijith et al., 2017). Studies have shown that green roofs can substantially sequester carbon in plants through photosynthesis reducing ambient CO_2 concentrations (Rowe, 2011).

Need for an Integrated Approach

Cities have a key role in resolving the sustainability challenges which have to be addressed through all the three pillars of sustainability-(Social) people, planet (Environmental) and profit (Economic). An integrated framework should be in place for assessing exposure, based on the atmospheric environmental quality of the cities and to identify vulnerable areas. This would require an understanding of the spatial and temporal distribution of thermal stress and air pollution in the city. It has been reported that India a has high potential for co-benefits of mitigation of air pollution and climate change. However, the knowledge on the interplay between these variables is still unclear. With intricate linkages between urban climate and air quality, it becomes important to understand their integrated exposure which could be of great interest for researchers in both health and climate studies. From practitioners' perspectives, such data could be of great help for urban planners in planning and designing sustainable cities.

Proposed Framework for Implementing NbS in Urban Areas

Identifying priority areas that require immediate attention is the major and foremost step. Both UHI and air pollution are reported to exhibit spatial as well as temporal (diurnal and seasonal) variations in urban areas. The large temperature variations in urban areas were attributed to various anthropogenic activities such as industries, transportation, air-conditioning and the urban design/geometry leading to changes in land use land cover (Gogoi et al., 2019). Air pollutants show large spatial and temporal variability depending on pollution sources, meteorology, microclimate and other factors. Hence hotspot mapping of air

TABLE 2 | Air pollution tolerance and sensitivity of commonly studied plant species in Indian cities.

Plant species	APTI value	Use	Location	Reference	
		Urban areas			
Mangifera indica	High	AP mitigation, Green belt development	Allahabad, UP	Kuddus et al. (2011)	
			Nanded, Maharashtra	Yannawar (2014)	
			Varanasi, UP	Pandey et al. (2016)	
Azadirachta indica	High to moderate	AP mitigation, High dust deposition due to good	Allahabad, Gujarat Nanded, Maharashtra	Kuddus et al. (2011) Yannawar (2014)	
		foliage	Ahmedabad, Gujarat	Chaudhary and Rathore (2019	
Eucalyptus	Moderate to sensitive	Can be used as bioindicator	Nanded, Maharashtra	Yannawar (2014)	
			Haridwar, Uttarakhand	Swami and Chauhan (2015)	
Ficus religiosa	High	Hardwar, UttarakhandModerate dust removal capacity,Allahabad, UPGreen belt developmentNanded, MaharashtraExcellent performer based on APIGandhinagar, Gujarat(anticipated pollution index)Varanasi, UP		Kuddus et al. (2011) Yannawar (2014) Chaudhary and Rathore (20 Pathak et al. (2011) Pandey et al. (2015a)	
			Ahmedabad, Gujarat	Chaudhary and Rathore (2019	
Dalbergia sissoo	Moderate	High dust removal capacity	Gandhinagar, Gujarat	Chaudhary and Rathore (2018	
		Industrial areas			
Mangifera indica	Moderate	Good dust capturing capacity, Phytoextractor of copper	Jamshedpur, Jharkhand	Roy et al. (2020)	
Azadirachta indica	Moderate	Tolerant to AP	Jamshedpur, Jharkhand	Roy et al. (2020)	
Ficus religiosa	High	Tolerant to AP, Good dust capturing	Talkatora Industrial Area, UP	Bharti et al. (2018)	
		capacity, Phytoextractor of copper	Jamshedpur, Jharkhand	Roy et al. (2020)	
Eucalyptus globus	High	Tolerant to AP	Talkatora Industrial Area, UP	Bharti et al. (2018)	
Bougainvillea sp	High	Tolerant to AP	Manali, Chennai	Ranjan et al. (2015)	
Shorea robusta	High	Tolerant to AP and good metal accumulation capacity	Bankura, West Bengal	Karmakar and Padhy (2019)	

pollution also becomes imperative for understanding its spatial and temporal patterns and to identify Poor Air Quality (PAQ) pockets within the urban area. Spatio-temporal UHI maps (urban and non-urban pockets delineated using Local Climate Zones [LCZ]) and air quality maps (spatial distribution of air pollution) will help in identifying city hotspots for combined exposure to thermal stress and air pollution. In order to design appropriate adaptation strategies, it is essential to further the understanding of the city's vulnerabilities to the risks under consideration. Physiological heat stress affects productivity which could further decrease the adaptive capacity of vulnerable population. Hence, the vulnerable areas also need to be identified based on the socio-economic characteristics of the population. The selection of potential implementation areas and NbS in the identified priority location should go hand in hand and requires detailed planning (Albert et al., 2020). Various stakeholders including researchers, urban planners, ecologists and social scientists should be actively involved in the planning stage as the successful implementation depends on the landscape and land use characteristics, cost-effectiveness, funding, other societal and practical challenges as depicted in Figure 2.

The selection of NbS strategy mainly depends on the location characteristics such as major air pollution sources, urban/street morphology, aesthetic use and other benefits such as erosion prevention, noise mitigation, economic value etc. To start with, a database of trees for a particular city has to be developed which could be further classified based on tree physiological characteristics, APTI value and thermal performance. This will help in identifying and designing tree forms to achieve the required reduction in pollution and urban heat.

Air Pollution Characteristics

The plant species for green space development can be selected from the database based on the air pollution characteristics at the location. For example, if industrial pollution is prominent in the location, then pollutant-specific tolerant species can be selected for the development of the green belt. To curb vehicular pollution along the roads, the plants with good dust capturing potential and canopy could be selected along with high pollution tolerance. Vegetation can also be designed as a barrier to protect and separate people from pollution (Abhijith



et al., 2017; Barwise and Kumar, 2020). However, vegetation can sometimes hinder dispersion, thus building up pollutant concentration (Janhäll, 2015). A careful selection of plant species and design parameters as well as meteorological conditions play a major role in the successful mitigation of air pollution in urban areas (Leung et al., 2011).

Thermal Reduction Potential

The thermal performance of the selected plant species should be evaluated to determine the heat reduction potential (HRP) or cooling potential. This could be determined based on various indicators such as air temperature, thermal comfort or surface temperature. A recent study evaluated a methodological framework to select the trees for urban heat mitigation through parametric ENVI-met simulations using thermal comfort as an indicator for assessing the cooling potential of trees (Morakinyo et al., 2020). The study results indicated that a significant reduction in urban heat can be achieved by selecting right tree at the right place.

Urban Morphology

Urban morphology also has a greater role in the successful implementation of NbS strategies. The type of NbS strategy that could be adopted largely depends on the configuration of the urban area i.e., land use, available space, street configuration etc. As mentioned above, tall trees/vegetation is not recommended in street canyons as it can hinder free flow of air thus increasing air pollution and temperature (Janhäll, 2015). At the same time, properly designed green spaces could effectively mitigate UHI by improving natural cooling and, air pollution. In case of space constraints, other strategies such as VGS, green roofing, moss wall (a wall covered with moss) could be implemented. The reclamation of wastelands, landfills and dump yards in the form of parks/gardens are also commonly practiced widely in urban areas. Some of the examples from India include cities such as Delhi, Kolkata, and Bangalore.

Longevity/Sustainability

As vegetation is highly influenced by environmental stress, climate and various other factors such as soil type, water requirement etc, the suitability of plant species to each site determines its longevity (Barwise and Kumar, 2020). The native plant species exhibiting tolerance to environmental stresses experienced by the site as well as other factors as discussed above ensures sustainability and effectiveness of NbS strategies.

Barriers in NbS Implementation

Even though there has been an increased interest in NbS based approaches in recent years with a growing number of studies indicating its co-benefits, it is currently lacking actions at the implementation level. Governance plays a major role in the

successful implementation of NbS (Li et al., 2021). Due to its multifunctionality, a participatory process is required with multi-stakeholder involvement and proper financial mechanisms to fund such approaches. In rapidly urbanizing nations like India, inadequate financial resources and space constraints are identified as major factors hindering its implementation (Singh et al., 2020a). The rapid developmental projects put immense pressure on cities in terms of space availability compromising the green spaces in urban areas. A recent report by UNEP estimated that globally, ~USD 133 billion/year is currently invested into NbS (using 2020 as the base year), which is considerably less than that flows to climate finance (United Nations Environment Programme, 2021). For countries like India, internationally comparable data on NbS financing is not available. The nationallevel policies in India on air pollution and climate change have individually allocated some funds for greening activities (under NCAP, INR 60.5 million is allocated for greening activities in 28 identified non-attainment cities), but lacks strategic planning. As NbS strategies are largely dependent on the local knowledge as well as socio-political conditions, adaptive governance is required based on the local knowledge and culture. This limits the decision-makers and funders from investing in this approach as they are not confident on its success and scalability. To address this challenge, recently the International Union for Conservation of Nature (IUCN) has established NbS global standard and self-assessment tool that will help to assess the effectiveness of NbS approaches (IUCN, 2020). IUCN has established a governance structure through an International Standard Committee and multistakeholder involvement. This will help in the effective mainstreaming and upscaling of NbS approaches into developmental planning and policies.

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CONCLUSION

NbS have great potential in mitigating the harmful impacts of climate change and air pollution. However, to maximize the potential of NbS for combined mitigation of UHI and air pollution, proper planning in the implementation of these strategies is needed. NbS should be adapted to local conditions to be successful and effective. Recently greening strategies have been increasingly promoted through various schemes and programs on air pollution and climate change such as the National Mission for Green India as a part of the National Action Plan on Climate Change (NAPCC) and city clean air action plans and smart city plans, but lack a strategic approach to green space planning and management. Also, there is a need for developing strong evidence base on the effectiveness of NbS, its co-benefits, and successful integration of green infrastructures into the city planning process, especially for a rapidly urbanizing country like India. Equitable access to green spaces has to be ensured for the sustainable and inclusive growth of cities. For example, socially disadvantaged groups are more prone to the negative impacts of air pollution and urban heat and likely to live in less green neighborhoods. Hence, NbS has to be integrated into the urban planning agenda providing a systematic approach including all the relevant stakeholders especially the citizens.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct and intellectual contribution to the work, and approved it for publication.

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Particulate Matter Pollution in Urban Cities of India During Unusually Restricted Anthropogenic Activities

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The outbreak of COVID-19 is a global public health challenge and has affected many countries, including India. The nationwide lockdown was imposed in India from March 25 to May 31, 2020 to prevent the transmission of COVID-19. The study intends to assess the impact of the absence of major anthropogenic activities during the various phases of the COVID-19 lockdown (LDN) period on the daily mean concentrations of PM_{2.5} and PM₁₀ in six populated cities of Jaipur, Jodhpur, Kota, Udaipur, Ajmer, and Alwar in the state of Rajasthan. Investigation has been done for the different periods, including the pre-lockdown-PRELD (January 1-March 4, 2020), partial lockdown-PLDN (March 5-24, 2020), COVID-19 lockdown-LDN (March 25-May 31, 2020), and unlocking-ULC (June 1-August 31, 2020) phases. We have also compared the mean concentrations of $PM_{2.5}$ and PM_{10} with the same period of the year 2019. A significant improvement in air quality during the COVID-19 LDN period was noticed in all cities compared to 2019 and for the same period of the year 2020. However, the levels of $PM_{2.5}$ and PM_{10} were seen to rise during the second, third, and fourth LDN phases compared to the first LDN, indicating that the subsequent lockdowns started with some relaxations and dusty conditions. On the other hand, wind-blown dust is another vital source of PM₁₀, resulting in high concentrations in the summer months (April–May). Significant reductions in PM2.5 (~25–50%) and PM10 (20–37%) in all six cities during the LDN period compared with PRELD were estimated. However, with significant variations from city to city, the lowest reductions in $PM_{2.5}$ (~25%) and PM_{10} (~20%) were measured in Jodhpur and Ajmer, respectively. It was noticed that the episodes of rainfall and transport of oceanic air masses resulted in a reduction of particles during the ULC period compared to the LDN period. The air quality index was, more or less, in the "good to satisfactory" category during the first 3 LDN periods, whereas it was moderate for Jodhpur, Jaipur, and Ajmer during the last LDN period. The study will be helpful to determine mitigation policies to minimize air pollution, especially in developing regions.

Keywords: particulate matter, COVID-19 lockdown, air quality index, meteorology, India

HIGHLIGHTS

- The investigation shows the impact of the various phases of COVID-19 lockdown on $PM_{2.5}$ and PM_{10} in six cities of Rajasthan.
- There was a reduction during the COVID-19 lockdown, by 16–50% in $PM_{2.5}$ and by 22–47% in PM_{10} .
- Mineral dust pollution is prominent in particle loads over the Rajasthan state.
- Monsoon rains and winds dominate over emissions during the unlocking periods (June–August) in $PM_{2.5}$ and PM_{10} distribution.

INTRODUCTION

The entire world has been facing an acute pandemic and challenging circumstances because of the novel contamination of coronavirus disease (COVID-19) since the beginning of 2020. The first case was identified in Wuhan, China, in December 2019 (Li et al., 2020). In view of the expanding pace of COVID-19 cases worldwide, the transmittable disease has become a global pandemic (WHO, 2020). Therefore, many countries have declared a nationwide strict lockdown with varying rules and regulations to control the spread of COVID-19. In India, the first COVID-19 case was identified on January 30, 2020 in Trisharu, Kerala (India) (WHO, 2020) and in different parts of the country during the first week of March (https://www.mohfw.gov.in/). The multiple cases of infection from India started to come into view in the first week of March 2020. Later, the government of India (Prime Minister) announced "Janata Curfew" for a

day on March 22, 2020, with a complete shutdown of daily activities. Later, complete lockdown for 21 days across India was officially announced, from March 25 to April 14, 2020. Later, to deal with the decline of the pandemic in the nation, the lockdown was extended up to May 3, 2020. After looking at the problematic situation, it continued until May 31, 2020 with some relaxation. The details of prohibited and permitted activities during the different lockdown phases, including LDN1 (March 25-April 14, 2020), LDN2 (April 15-May 3, 2020), LDN3 (May 04-17, 2020), LDN4 (May 18-31, 2020), are listed in Table 1. Improvements in air quality due to reductions in transport (flights, trains, and vehicles), industrial, academic, economic, and social-related activities are reported in several studies (Dantas et al., 2020; Kumari and Toshniwal, 2020; Nakada and Urban, 2020; Navinya et al., 2020; PPAC, 2020; Sharma et al., 2020; Sicard et al., 2020; Singh et al., 2020; Yadav et al., 2020). About 40-70% reductions were suggested in transport, industrial, and construction activities based on the fuel consumption data during the lockdown period, but ~12% enhancement was estimated in biofuel consumption for cooking purposes (PPAC, 2020). The combustion-related emissions are considered one of the significant sources of air pollutants in Indian cities. The air quality, in terms of air quality index and health impact, is mainly determined by the levels of hazardous particulate pollutants in a year. In ambient air, the most important anthropogenic sources of fine particulate matter are the incomplete combustion of biofuel and fossil fuels (Sahu et al., 2011). In addition to local emissions, the long-range transport from the regions of biomass burning and dust sources also influence the receptor sites (Yadav et al., 2016, 2017). Exposure to elevated levels

TABLE 1	Details of the prohibited and permitted activities during the study period.	

Study phases and its duration/2020	Total days	Activities
PRELD January 1–March 4	63	Business as usual
PLDN March 5–24	21	Selective restrictions announced by the government, like mass gatherings in institutions, shopping malls, and theaters
LDN1 March 25–April 14	21	All were closed, with the exception of essential services like for health. Security and food delivery and others
LDN2 April 15–May 3, 2020	19	Allowed were agricultural businesses including dairy, banking, telecommunication, power-related services, manufacturing, and transportation units; interstate movement was also allowed for the delivery of essential goods and for law and order
LDN3 May 4–17, 2020	14	Allowed were all activities but with restrictions and social distancing, e.g., travel by air and rail around the metro running of schools, colleges, and other educational and training/coaching institutions; hospitality services, including hotels and restaurants; places of large public gatherings, such as cinema halls and malls Gymnasiums, sports complexes, etc., were still closed
LDN4 May 18–31, 2020	14	Protection for vulnerable persons, i.e., persons above 65 years of age, persons with co-morbidities, pregnant women, and children below the age of 10 years shall stay at home, except for meeting essential requirements and for health purposes
ULC1 June 1–30, 2020	30	Shopping malls, religious places, hotels, and restaurants were permitted to reopen from June 8. Large gatherings were still banned, but there were no restrictions on interstate travel
ULC2 July 1–31, 2020	31	Shops were permitted to allow more than five persons at a time. Educational institutions, metros, and recreation centers remained close until July 31.
ULC3 August 1–31, 2020	31	Educational institutions would remain close until August 31. All inter- and intrastate travel and transportation modes are permitted. Independence Day celebrations are permitted with social distancing

MHA, 2020.

of PM_{2.5} in ambient air can cause adverse health effects like respiratory and cardiovascular diseases (e.g., Pope et al., 2009). Several studies have reported the association of COVID-19related mortality and morbidity along with air pollution and meteorological parameters during the lockdown period (Beig et al., 2020; Conticini et al., 2020; Wu et al., 2020; Yao et al., 2020). On the other hand, many studies have reported a significant reduction in air pollution across the world due to the COVID-19 restrictions (Bashir et al., 2020; Jain and Sharma, 2020; Kanniah et al., 2020; Kumar et al., 2020; Kumari and Toshniwal, 2020; Mahato et al., 2020; Nakada and Urban, 2020; Navinya et al., 2020; Singh and Chauhan, 2020; Singh et al., 2020; Tobías et al., 2020; Yadav et al., 2020; Goel et al., 2021; Sokhi et al., 2021). Most of the studies in India have been focused on megacities so far, including Delhi, Mumbai, Ahmedabad, Pune, Chennai, Kolkata, Lucknow, Jaipur, and Bangalore, etc. However, studies during the COVID-19 lockdown period in several cities of Rajasthan are reported for a very limited period (Sharma et al., 2020). Aside from local anthropogenic emissions, the air pollution footprints are unique in Rajasthan, where dust-related sources are also dominant. The geographic characteristics of Rajasthan are the Thar Desert and the Aravalli Range, and the north-western portion of Rajasthan is generally sandy and dry. Hence, due to being the driest region and given the different climatic zones, the study will be very prominent in these locations. The present study highlights the role of long-term COVID-19 lockdown on PM_{2.5} and PM₁₀ in six major cities of Rajasthan, including Jaipur, Jodhpur, Kota, Udaipur, Ajmer, and Alwar, and the alteration in air pollution levels during the unlocking periods. We have also discussed the change in the air quality index (AQI) for all cities. Overall, we present a detailed investigation by considering the combined effects of local emissions and meteorology during the lockdown period.

METHODOLOGY

Data Sources, Locations, and Local Meteorology

The concentration data of $PM_{2.5}$ and PM_{10} as measured in six cities, namely, Jaipur, Jodhpur, Kota, Udaipur, Ajmer, and Alwar, in the state of Rajasthan were obtained from the Central Pollution Control Board (CPCB) for the years 2019 and 2020. The $PM_{2.5}$ and PM_{10} data in these cities located in the northwest part of India were analyzed to investigate the pollution levels during the long-term COVID-19 lockdown period. The locations of the study sites in the maps of India and Rajasthan are shown in **Figure 1**.

The data studied include during the pre-lockdown period (PRELD) (January 1 to March 4, 2020), partial lockdown (PLDN) (March 5 to 24, 2020), COVID-19 lockdown (LDN) (March 25 to May 31, 2020), and unlocking (ULC) period (June 1 to August 31, 2020). The average matching periods of the year 2019 were considered as the references to study the change in air quality during the COVID-19 lockdown period. We have used the National Centre for Environmental Prediction reanalysis datasets to derive the wind streamlines



and temperature at a spatial resolution of $2.5^{\circ} \times 2.5^{\circ}$ during the entire study period for 2019 and 2020, as shown in **Figure 2**. The precipitation rate (mm day⁻¹) data from the CPC Merged Analysis of Precipitation (CMAP), at a spatial resolution of $2.5^{\circ} \times 2.5^{\circ}$, was used. The Moderate Resolution Imaging Spectroradiometer (MODIS) active fire count maps over India during the entire study period for 2019 and 2020 are plotted in **Figure 3**. Rajasthan is the largest state of India, located in the north-western region with a population of ~6.8 million (http://census2011.co.in). However, being located near most cities, the state has various types of industrial units, including those of marble, steel, cement, handicrafts, textiles, chemicals, IT, and thermal plants (Report 2018). The primary sources of air pollutants are vehicular, industrial, agriculture residual burning, municipal solid waste treatment,



FIGURE 2 | Meteorological parameters during the different phases of the study in the years 2019 and 2020: (top) wind speed; (middle) temperature; (bottom) precipitation.



diesel generator, biofuel and LPG (residential cooking), and construction activities. The common sources of $PM_{2.5}$ and PM_{10} include biofuel and fossil fuel combustion, industrial processes, and biomass burning. However, for PM_{10} , wind-blown dust, road dust suspension, building construction, lime kilns, and slab polishing are important non-combustion sources (Yadav et al., 2017). Daily mean temperature ranges of 10–27, 32–45, and 35–40°C are recorded during winter (January–February), pre-monsoon (March–May), and monsoon (June–September) seasons, respectively. The strong hot winds called "Loo" blow from the west during pre-monsoon, while relatively low winds blow from the north and northeast during the winter (Yadav et al., 2014a). Rainfall values as high as 819 mm in the eastern part and as low as 170 mm in the western part of the state were recorded (Maanju and Saha, 2013). In the monsoon season, the

inter-tropical convergence zone moves northward across India, and strong southwest (SW) winds prevail over the study region. Therefore, the measurements in this season are influenced by the transport of cleaner air masses from the Arabian Sea (see **Supplementary Figure S1**).

RESULTS AND DISCUSSION

Role of COVID-19 Lockdown in the Variation of PM_{2.5} and PM₁₀

The time series analysis of $PM_{2.5}$ and PM_{10} over a period of 8 months (January 1–August 31, 2020) reveals interesting trends. The daily means of $PM_{2.5}$ and PM_{10} concentrations measured in Jaipur, Jodhpur, Kota, Udaipur, Ajmer, and Alwar are shown in **Figure 4**. The daily mean time series concentrations



of PM_{2.5} and PM₁₀ for all cities are divided into the four different periods of PRELD (January 1–March 4), PLDN (March 5–24), COVID-19 LDN (March 25–May 31), and ULC (June 1–August 31). During the business-as-usual period, many studies have reported a clear seasonality in the daily mean concentrations of PM_{2.5} and PM₁₀, with the highest in the dry period (winter to pre-monsoon) and the lowest in the monsoon season, in different cities of India (Beig et al., 2007; Yadav et al., 2017; Hama et al., 2020). However, the findings become effectively more crucial in 2020 to researchers and policymakers because of the COVID-19-related consecutive LDN and ULC situations in which anthropogenic activities were restricted. The daily mean concentrations of PM_{2.5} were in the ranges of 7–81, 18– 156, 12–91, 14–93, 10–114, and 15–63 μ g m⁻³, while those of PM_{10} varied in the ranges of 20–256, 39–282, 23–185, 26–202, 15–266, and 30–121 μg m $^{-3}$, respectively, in Jaipur, Jodhpur, Kota, Udaipur, Ajmer, and Alwar during the entire study period. The mean concentrations of $PM_{2.5}$ were 51 \pm 10, 78 \pm 21, 45 \pm 13, 49 \pm 12, 47.3 \pm 13, and 47.2 \pm 8 μg m $^{-3}$ in the PRELD period, which reduced to 34 \pm 09, 58 \pm 16, 29 \pm 3, 29 \pm 2, 32 \pm 11, and 24 \pm 4.4 μg m $^{-3}$ during the LDN period in Jaipur, Jodhpur, Kota, Udaipur, Ajmer, and Alwar, respectively. On the other hand, the mean concentrations of PM₁₀ were 115 \pm 23, 162 \pm 41, 97 \pm 25, 104 \pm 18, 100 \pm 25, and 85 \pm 9 μg m $^{-3}$ in the PRELD period, which decreased by 90 \pm 27, 116 \pm 29, 67 \pm 8, 65 \pm 14, 80 \pm 35, and 53 \pm 10 μg m $^{-3}$ during the LDN periods in Jaipur, Jodhpur, Kota, Udaipur, and Alwar, respectively. The

TABLE 2 | The daily average mass concentrations of PM_{2.5} and PM₁₀ during different periods, namely, PRELD, PLDN, and LDN phases and unlocking phases in six different cities of Rajasthan in India.

Cities	PRELD	PLDN	LDN1	LDN2	LDN3	LDN4	LDN periods	ULC1	ULC2	ULC3	ULC periods
PM _{2.5}											
Jaipur	50.8 ± 10	39.9 ± 12	23.3 ± 8	30.8 ± 11	36.8 ± 12	46.7 ± 8.4	34.4 ± 9.8	28.3 ± 7.1	19.9 ± 8.2	14.9 ± 5.5	21.0 ± 6.7
Jodhpur	78 ± 21	69.6 ± 23	47.8 ± 14	46 ± 11	57.7 ± 23.5	82.3 ± 30	58 ± 16	54.6 ± 14.7	51.7 ± 6.9	42.1 ± 6.5	49.5 ± 6.5
Kota	45.7 ± 13	38 ± 9	27.7 ± 5.3	28 ± 5	28.4 ± 5.3	33 ± 6.5	29 ± 3	22.4 ± 4	18.7 ± 4	21.8 ± 3.9	21 ± 1.9
Udaipur	49 ± 12	36 ± 7	26 ± 6	25 ± 5	31 ± 7	34 ± 11	29.3 ± 2.5	26.8 ± 5	25.1 ± 4.5	23.6 ± 5	25 ± 1.6
Ajmer	47 ± 13	49 ± 18	22 ± 5	24.7 ± 4.2	33.5 ± 10	47.7 ± 16	32.2 ± 11	31.7 ± 7.8	26.4 ± 6.6	23.5 ± 5	27.2 ± 4
Alwar	47 ± 8.2	41.6 ± 3.4	23 ± 5	20.6 ± 3	23.4 ± 3.8	32.5 ± 5	24 ± 4.4	39 ± 7	38 ± 7.2	28.7 ± 3.3	35.3 ± 5.7
PM10											
Jaipur	115 ± 23	96.6 ± 46	60.5 ± 24	81.3 ± 44	94 ± 41	125 ± 38	90.37 ± 27	74 ± 31	50.7 ± 23	36.5 ± 12	54 ± 19
Jodhpur	162 ± 41	154 ± 53	96.2 ± 26	96 ± 19	114 ± 40	159 ± 28	116 ± 29	113 ± 28	103 ± 15.3	89 ± 16	101 ± 12
Kota	96.6 ± 25	88 ± 20	69.8 ± 14.9	63 ± 13.9	59 ± 9	76 ± 19.9	67 ± 8	47.9 ± 10.6	37.5 ± 8.8	39.6±7	42 ± 5
Udaipur	103.6 ± 18.8	76.5 ± 19	54.7 ± 14	59 ± 22.6	60.5 ± 12	87.4 ± 47	65 ± 14	55.4 ± 14	53 ± 10.9	48.9 ± 15	52 ± 3
Ajmer	99.9 ± 25	103 ± 35	55.7 ± 18	60.6 ± 12.6	71.9 ± 20.9	131.6 ± 57	80 ± 25	72 ± 24	54 ± 14	49 ± 12	58 ± 12
Alwar	84.5 ± 9.9	83.4 ± 11	48.7 ± 15	41 ± 7.9	56 ± 13.2	65.3 ± 12	53 ± 10	75.9 ± 16	68.5 ± 10	54 ± 5.5	66 ± 11

PRELD, pre-lockdown; PLDN, partial lockdown; LDN, lockdown; ULC, unlocking.

daily mean concentrations of PM_{2.5} and PM₁₀ during PRELD, PLDN, various phases of LDN, and UCL situations are shown in Table 2. The National Ambient Air Quality Standards (NAAQS) recommend the daily permissible $PM_{2.5}$ of 60 $\mu g m^{-3}$ and PM_{10} of 100 µg m⁻³ for India (MoEFCC, 2015). About 3, 33, and 6% of PM2.5 daily data exceeded the permissible limit of NAAQS during the COVID-19 LDN period in Jaipur, Jodhpur, and Ajmer, while 35, 46, 4, 7, and 19% of PM₁₀ exceeded in Jaipur, Jodhpur, Kota, Udaipur, and Ajmer, respectively. It is suggested that the concentrations of PM2.5 significantly decreased in all cities that met the NAAQS limits. However, in a few cities, like Jodhpur, Jaipur, and Ajmer, the PM₁₀ levels did not fall below the standard limit during most of the COVID-19 LDN period due to non-combustion-related sources like wind-blown dust. The highest reduction by 47% in PM_{2.5} was observed in Alwar, followed by Udaipur (40%), Kota (36%), Jaipur (32%), and Ajmer (32%), while Jodhpur showed the lowest reduction of about 25% during the COVID-19 LDN, respectively. PM₁₀ was reduced by 37% in Alwar and 36% in Udaipur. However, the lowest reductions of 19-30% were estimated for Jaipur, Jodhpur, Kota, and Ajmer cities (Figure 5). Additionally, we have compared the $PM_{2.5}$ and PM_{10} concentrations measured in 2020 with those during the previous year (2019) for the COVID-19 LDN and ULC periods (see Figure 6). The differences are, more or less, the same during PLDN, but significantly lower concentrations of both pollutants were estimated during the COVID-19 LDN and ULC periods. Wilcoxon test has been performed, and P-value < 0.05 was found, which was considered statistically significant in the LDN periods. The reductions in PM_{2.5} and PM₁₀ varied in the ranges of 16-50 and 22-47% during the COVID-19 LDN period with respect to the same period of the year 2019 (without meteorological normalization), as shown in Figure 7. The reductions of 6-46% in PM_{2.5} and 28-49% in PM₁₀ were estimated during the overall ULC period.

Overall, the declining trends of PM_{2.5} and PM₁₀ observed in all cities during the COVID-19 LDN period are attributed to the reduction in combustion-related emissions (NASA, 2020; Navinya et al., 2020). Conversely, to some extent, elevated levels of PM2.5 and PM10 were observed during the LDN3 and LDN4 periods (see Figure 4). During all COVID-19 LDN phases, the restrictions were diverse and resulted in variations of PM2.5 and PM₁₀ in each LDN phase—for example, major anthropogenic and industrial activities were stopped, except for emergency services like the health sector (Yadav et al., 2020). Later, some relaxations with limitations were allowed in the subsequent LDN periods (MHA, 2020). Sahu et al. (2011) have reported that the emissions from transport and industrial sectors have significant contributions to the ambient concentrations of both PM2.5 and PM₁₀. In addition to this, wind-blown dust is a more substantial source of coarser particles (PM_{10}) in the northern and western regions of India (Sahu et al., 2011; Yadav et al., 2014b). During the ULC period, only Alwar City exhibited higher values of PM_{2.5} and PM₁₀ compared with the overall LDN phases in the same year; this could have happened due to limited local rainfall events. Interestingly, the highest plummet can be attributed to rainfall in the ULC period (monsoon) compared to 2019. In the monsoon period, episodes of rainfall can wash out the dust particles (PM₁₀) much faster than the finer particles (PM_{2.5}), unless there is frequent rainfall for many days (Yadav et al., 2014c). Figure 8 shows the ratios of $PM_{2.5}/PM_{10}$ during the study period in the year 2020 in six cities that were used to investigate aerosol combustion and non-combustion sources. The ratio of $PM_{2.5}/PM_{10}$ was observed to be <0.50 µg µg⁻¹ in all cities due to higher temperatures during the COVID-19 LDN period (premonsoon months), leading to dry and dusty conditions (Chan and Yao, 2008). The emissions from wind-blown dust are highly significant, mainly contributing to PM₁₀, and are probably the reason for the high levels of coarser particles. Interestingly, about 0.40 μ g μ g⁻¹ ratio of PM_{2.5}/PM₁₀ was observed during all phases



of COVID-19 LDN in Jaipur City (nearby Delhi) due to the impact of dust storm. In the pre-monsoon season, dust storms originate from Arab countries, and the Thar Desert can influence most regions, like Delhi, Rajasthan, etc., of India (Anand et al., 2019), thus rapidly increasing the PM_{10} levels. Overall, dust pollution is prominent in particle loads over the Rajasthan state. In addition to this, we have incorporated the plots of O₃/NO₂ and SO₂/NO₂ ratios during the study period in **Figure 8**. In the LDN periods (pre-monsoon months), the high O₃/NO₂ ratios were due to the transport of photochemical pollutants

from higher altitudes. The efficient vertical mixing with free tropospheric air increases O_3 in the pre-monsoon months. The enhanced SO_2/NO_2 indicates that the predominant source of SO_2 is coal-based thermal power plants. In other words, the site receives reasonably aged SO_2 from distant regions, resulting in high ratios (Mor et al., 2021).

Impact of Meteorology and Biomass Burning on Air Quality

In addition to the local emissions, the variation in ambient air pollutants can also be influenced by the local meteorology, transport, transformation, and scavenging by the precipitation (Sahu et al., 2016a,b, 2020; Yadav et al., 2019a,b). Therefore, the mean wind streamlines along with temperature and precipitation at 925 mb pressure level are used in the study to investigate any unusual changes in meteorological and transport processes during the COVID-19 LDN period. The comparison of meteorological parameters during 2020 and 2019 for the PRELD, COVID-19 LDN, and UCL periods is shown in Figure 2. The trends in wind speed, temperature, and precipitation indicate the different features during the entire study period due to the change of seasons from winter to pre-monsoon and then monsoon (Yadav et al., 2014a; Sahu et al., 2017). Hence, the trends of PM2.5 and PM10 may also include a factor related to the change of meteorological conditions in the same year in comparison with the PRELD period. In the pre-monsoon season (LDN period), high temperatures, high wind speeds, and more profound boundary layer heights may lead to a higher dispersion of pollutants and lower concentrations (Sahu et al., 2020). However, the difference between 2019 and 2020 does not show significant fluctuations in wind speed, temperature, and precipitation in the western region of India during the PRELD to ULC periods. The mean surface temperatures and rainfall were in ranges of 30-32°C and 0-1.5 mm day⁻¹ during the COVID-19 LDN period in the years 2019 and 2020 over the study region. The strong westerly wind prevails over the area during both years in the COVID-19 LDN period. In general, all meteorological parameters were, more or less, similar during the study period, especially in the COVID-19 lockdown period in 2019 and 2020. Several studies have reported that the meteorological parameters were identical in the previous years and 2020 during the COVID-19 LDN over the Indian region (Navinya et al., 2020; Sharma et al., 2020; Singh et al., 2020; Yadav et al., 2020). Additionally, in the pre-monsoon season, the local emissions become lesser, but outside contributions become more prominent due to the increased transport of regional air masses. Furthermore, \sim 7% open agricultural burning in this season is mainly due to the burning of residues from wheat crops. Figure 3 shows MODIS-active fire maps over India during the entire study period for the years 2019 and 2020. In addition to this, to some extent, the SW winds also influenced the region after passing through populated areas of central India; they may get polluted during the pre-monsoonto-monsoon transition period (Sahu et al., 2020; Maji et al., 2021; Yadav et al., 2021). The maps clearly show that the premonsoon fire events in 2020 during COVID-19 LDN have



somewhat decreased to a shallow level compared to those in 2019, especially in western, northern, and central India (Kant et al., bib2020), as biomass burning is a significant aerosol source. Overall, the reductions in fire events along with local emissions have also been deficient. Additionally, we have normalized the mass concentrations of $PM_{2.5}$ and PM_{10} with meteorology to investigate the actual change in emissions due to COVID-19

LDN. In the method, ventilation coefficient, a product of wind speed (WS) and boundary layer height (BLH), is used to see the role of weather dynamics for all cities of Rajasthan. The WS (above 10 m) and BLH datasets used in the study are from Copernicus Emergency Management Service (Climate Data Store) (https://cds.climate.copernicus.eu/cdsapp#!/search?type= dataset). The extensive details of normalizing the meteorological



effects on ambient pollutants are reported elsewhere (Dai et al., 2020; Falocchi et al., 2021; Mishra et al., 2021). The reductions in PM_{2.5} and PM₁₀ (with meteorology-normalized data) were in the ranges of 20–55 and 22–49% during the COVID-19 LDN period compared to the same period of 2019, as shown in **Figure 7**. Enhancement (differences ~10%) was observed in the reduction of PM_{2.5} and PM₁₀ (in meteorology-normalized data) compared to without meteorology-normalized data during the LDN period in all cities.

Air Quality Index in Rajasthan Cities

Air quality index is a tool for people to understand the air quality status in an easy way. This tool transforms the complex data of various pollutants into a single number (index value), classification, and color (Beig et al., 2010). AQI was calculated for all cities to identify the overall improvement in air quality, and the details of AQI are available elsewhere (CPCB, 2014). There are six AQI categories: good + satisfactory (0-100), moderately polluted (101-200), poor (201-200), very poor (301-400), and severe (401-500). We have converted the PM2.5, PM10, and SO₂ concentration values into AQI using typical values, as the minimum of three parameters was very much needed for the calculation. Therefore, only SO₂ concentration was used for the AQI analysis. Each category is decided based on the ambient concentration values of air pollutants and their likely health impacts known as health breakpoints. The AQ sub-index and breakpoint concentration of different pollutants are provided elsewhere (CPCB, 2014). Figure 9 indicates the AQI variation during the entire study period over six cities. Overall, the AQI was satisfactory to moderately polluted categories during the whole study period (PRELD to ULC). The AQI varied in the ranges of 85-161 during PRELD in all cities, with the highest in Jaipur, Jodhpur, and Udaipur cities, during which the traffic was regular. Later, the AQI declined during various LDN periods compared to during PRELD in all cities, except Jaipur, Jodhpur, and Ajmer. More or less, in all cities, increasing values were seen during the subsequent LDN periods but with the highest of 117 in Jaipur, 174 in Jodhpur, and 121 in Ajmer. Overall, in LDN1 to LDN3, all cities were in satisfactory ranges, but Jaipur, Jodhpur, and Ajmer showed moderate air quality conditions in LDN4. Interestingly, Jodhpur City gets good air quality during the third phase of ULC since the LDN periods. It suggests that more contributions to coarser particles (PM_{10}) are windblown dust and dust storms and, of course, anthropogenic activities, which have been allowed with limitations in the subsequent LDNs.

CONCLUSIONS

We have investigated the trends of the daily mean concentrations of $PM_{2.5}$ and PM_{10} in six cities, including Jaipur, Jodhpur, Kota, Udaipur, Ajmer, and Alwar of Rajasthan in India. We have obtained the data from CPCB and analyzed it for the years 2019 and 2020 during eight specific months (January to August). The primary aim is to understand the role of unusually low anthropogenic emissions in ambient concentrations of particulate pollutants during the COVID-19 lockdown. The investigation combines the effects of local anthropogenic emissions, meteorology, and the transport of biomass burning in the variations of both pollutants. The $PM_{2.5}$ and PM_{10} concentrations were reduced significantly in all cities and met the national standards during the lockdown. However, in a few cities, like Jodhpur, Jaipur, and Ajmer, the PM_{10} levels have not fallen below the standard



limit (100 μ g m⁻³) most of the time during the COVID-19 LDN period due to non-combustion-related sources like wind-blown dust. The highest reduction in PM_{2.5}, by 47%, was observed in Alwar, followed by Udaipur (40%), Kota (36%), Jaipur (32%), and Ajmer (32%), while Jodhpur showed the lowest reduction of about 25% during the COVID-19 LDN, respectively. PM₁₀ was reduced by 37 and 36% in Alwar and Udaipur. However, the lowest (19–30%) reductions were in Jaipur, Jodhpur, Kota, and Ajmer. More or less, there was 50% of $PM_{2.5}$ and PM_{10} in all cities due to higher temperatures during the COVID-19 LDN period (premonsoon months), supporting the uplifting of mineral and dust particles (PM10). In addition to the unusual local emissions, meteorological parameters, such as rainfall and movement of air masses, played a significant role in reducing the particle levels.



DATA AVAILABILITY STATEMENT

Publicly available datasets were analyzed in this study. This air quality data can be found form the CPCB (https://cpcb.nic.in/).

AUTHOR CONTRIBUTIONS

RY: conceptualization, software, and writing—original draft. PV: formal data analysis. PK and RY: data analysis. UP, PV, and VS: data collection. MG, PK, RY, and NT: methodology. PD, SJ, LS, GB, and DR: discussion. PD, SJ, LS, GB, DR, RY, PV, PK, UP, VS, MG, and NT: writing—review and editing. GB, RY, and LS: investigation. SJ, LS, and GB: visualization. All authors contributed to the article and approved the submitted version.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/frsc.2022. 792507/full#supplementary-material

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