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Comparative study of gaseous pollutants and particulate matter for major cities in the foothills of Garhwal Himalaya of Uttarakhand

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सार – इस शोध पत्र में, उत्तराखंड में गढ़वाल हिमालय के तीन प्रमुख शहरों (अर्थात् देहरादून, ऋषिकेश और हरिद्वार) में गैसीय प्रदूषकों (SO_2 और NO_2) और कणिकीय पदार्थ (PM_{10}) की परिवर्तनशीलता की जांच की गई है। उत्तराखंड प्रदूषण नियंत्रण बोर्ड द्वारा स्थापित पांच अलग-अलग रिकॉर्डिंग स्टेशनों के डेटा का उपयोग करके गैसीय प्रदूषकों और कणिकीय पदार्थ (2011-2020) के दशकीय व्यवहार का अध्ययन किया गया है। मौसम संबंधी प्राचलों के साथ SO_2, NO_2 और PM_{10} की मासिक और वार्षिक परिवर्तनशीलता का विस्तृत सांख्यिकीय विश्लेषण किया गया है। पांच अलग-अलग स्थानों पर, सीटी (क्लॉक टॉवर), आरआर (रायपुर रोड), आईएसबीटी (इंटर-स्टेट बस टर्मिनल), एनपीपी (नगर पालिका परिषद), और सिडकुल (राज्य औद्योगिक विकास निगम उत्तराखंड लिमिटेड) में SO_2 और NO_2 की सबसे कम मासिक सघनता क्रमशः 7.42, 6.73, 9.16, 5.98 और 3.03 g/m^3 और 8.93, 7.87, 11.63, 7.62 और 8.68 $\mu g/m^3$ थी। PM_{10} की सघनता CT, RR, ISBT, NPP और SIDCUL में क्रमशः 73.51-310.73 $\mu g/m^3$, 67.95-483.18 $\mu g/m^3$, 70.91-388.67 $\mu g/m^3$, 33.98-195.71 $\mu g/m^3$ और 46.08-220.64 $\mu g/m^3$ के बीच पाई गई। SO_2, NO_2 और PM_{10} की ऋतुनिष्ठ और वार्षिक परिवर्तनशीलता का भी विश्लेषण किया गया, और यह पाया गया कि मॉनसून ऋतु (वर्ष 2020 को छोड़कर) के दौरान गैसीय प्रदूषकों और कणिकीय पदार्थों की सघनता अपेक्षाकृत कम थी। गैसीय प्रदूषकों SO_2 और NO_2 की वार्षिक औसत सांद्रता CPCB (केंद्रीय प्रदूषण नियंत्रण बोर्ड) द्वारा निर्धारित NAAQS (राष्ट्रीय परिवेशी वायु गुणवत्ता मानक) की सीमा के अंदर थी, लेकिन NO_2 की सांद्रता WHO (विश्व स्वास्थ्य संगठन) के दिशानिर्देश मूल्यों से अधिक थी। PM_{10} की वार्षिक औसत सांद्रता CPCB द्वारा निर्धारित NAAQS और WHO द्वारा दी गई अनुमेय सीमा से बहुत अधिक थी। इस अध्ययन अवधि के दौरान मौसम संबंधी प्राचलों की परिवर्तनशीलता का भी विश्लेषण किया गया। गैसीय प्रदूषक SO_2 और NO_2 CT, RR और ISBT में क्रमशः 0.88, 0.93 और 0.94 के बीच एक बहुत मजबूत सकारात्मक सहसंबंध देखा गया है और PM_{10} का ISBT की साइट में SO_2 (0.62) और NO_2 (0.64) के साथ एक मजबूत सहसंबंध रहा है। सभी अध्ययन स्थलों पर गैसीय प्रदूषकों (SO_2 और NO_2) और कणिकीय पदार्थ (PM_{10}) के बीच रैखिक समाश्रयण प्लॉट दिखाए गए हैं। मौसम संबंधी प्राचलों के साथ वायु प्रदूषकों का वर्तमान विस्तृत विश्लेषण इस क्षेत्र में वायु गुणवत्ता नियंत्रण से संबंधित नीति-निर्धारण के लिए सहायक हो सकता है।

ABSTRACT. In the present paper, the variability of gaseous pollutants (SO_2 and NO_2) and particulate matter (PM_{10}) at three major cities (*viz*., Dehradun, Rishikesh and Haridwar) in the Garhwal Himalaya in Uttarakhand are investigated. The decadal behavior of gaseous pollutants and particulate matter (2011-2020) was studied by utilizing data from five different recording stations established by the Uttarakhand Pollution Control Board. A detailed statistical analysis of the monthly and annual variability of SO_2, NO_2 and PM_{10} along with the meteorological parameters is performed. At five different places, CT (Clock Tower), RR (Raipur Road), ISBT (Inter-State Bus Terminal), NPP (Nagar Palika Parishad) and SIDCUL (State Industrial Development Corporation of Uttarakhand Limited), the lowest monthly concentrations of SO_2 and NO_2 were 7.42, 6.73, 9.16, 5.98 and 3.03 g/m^3 and 8.93, 7.87, 11.63, 7.62 and 8.68 $\mu g/m^3$, respectively. The PM_{10} concentration was found to vary between 73.51-310.73 $\mu g/m^3$, 67.95-483.18 $\mu g/m^3$, 70.91-388.67 $\mu g/m^3$, 33.98-195.71 $\mu g/m^3$ and 46.08-220.64 $\mu g/m^3$at CT, RR, ISBT, NPP and SIDCUL respectively. The seasonal and annual variability of SO_2, NO_2 and PM_{10} were also analyzed and it was found that the concentration of gaseous pollutants and particulate matter was relatively lower during the monsoon season (except for the year 2020). The annual mean concentration of gaseous pollutants SO_2 and NO_2 was under the prescribed limit of the NAAQS (National Ambient Air Quality Standards) fixed by the CPCB (Central Pollution Control Board), but the NO_2 concentration was higher than the guideline values of the WHO (World Health Organization). The annual mean concentration of PM_{10} was much higher than the NAAQS by CPCB and the permissible limit given by WHO. The variability of meteorological parameters during the study period was also analyzed. A very strong positive correlation has been observed between the gaseous pollutants SO_2 and NO_2 (0.88, 0.93 and 0.94 in CT, RR and ISBT respectively) and PM_{10} have a strong correlation with SO_2 (0.62) and NO_2 (0.64) in the ISBT site. The linear regression plots between gaseous pollutants (SO_2 and NO_2) and particulate matter (PM_{10}) have been shown at all study sites. The present detailed analysis of air pollutants along with the meteorological parameters might be helpful for policy-making related to air quality control in this region.

Key words – Air pollution, Gaseous pollutants, Particulate matter, Himalayan foothills.

1. Introduction

Over the past few decades, air pollution has become one of the major environmental problems, especially in densely populated cities that challenge human health as well as a complete ecosystem (Pandey *et al*., 2015). The changes in physical and chemical characteristics of ambient air caused by either gaseous pollutants or suspended particles with detrimental impact on human health and the complete ecosystem are termed "air pollution" (Awasthi *et al*., 2016). Air pollutants can be both natural and anthropogenic and play a major role in altering the weather and climate of a particular geographical area (Deep *et al*., 2018; Mallik *et al.*, 2012). WHO (World Health Organization) estimates that air pollutants related to health issues are responsible for around seven million global deaths each year and nearly two million Indians died due to air pollution in 2019 (Lancet report 2020). Air pollutants may broadly be classified into two categories: primary and secondary. Pollutants directly released into the atmosphere from the emission source are called primary pollutants, whereas secondary pollutants are not directly emitted into the air but formed by the interaction or chemical reaction of primary pollutants (Shukla *et al.*, 2010; Saud *et al*., 2011). Nitrogen dioxide (NO_2) and sulphur dioxide (SO_2) are major primary pollutants having long-term deleterious impacts on human health and the environment (Joshi *et al*., 2011; Kuniyal *et al*., 2007). SO_2 and NO_2 are released into the urban environment, substantially from anthropogenic sources (Datta *et al*., 2010; Gavali *et al*., 2020). These gaseous pollutants predominantly come from factories, biomass burning, power plants, industries and vehicle emissions (Deep *et al*., 2017; Bhanarkar *et al.*, 2005; Kishore *et al*., 2017; Chauhan *et al*., 2010). These are the emissions of sulphur dioxide (SO_2) and nitrogen dioxide (NO_2), which have the potential to influence the chemical and radiation balance at the regional scale (Mallik *et al*., 2014). The burning of fossil fuels is also a major source of gaseous pollutants in the atmosphere and has a significant impact on the global chemical composition of the atmosphere and climate change (Sharma *et al*., 2010; Kishore *et al*., 2019). These pollutants are associated with many critical health issues, including respiratory problems, cardiovascular disease, lung cancer, asthmatics and chronic obstructive pulmonary disease (COPD), which is one of the major causes of global deaths (Dandotiya *et al*., 2020; Bralic *et al*., 2012; Agarwal *et al.*, 2006; Attri & Tyagi, 2010). The gaseous pollutants SO_2 and NO_2 are also responsible for acid rain formation, affecting the aquatic and terrestrial ecosystems (Behera *et al*., 2011; Sharma and Raina, 2012).

Particulate matter is a term for a mixture of solid and liquid in the air, that affects human health, climate and the ecosystems of nature (Ganguly *et al.*, 2019; Deep *et al*., 2017; Chauhan *et al*., 2010). The suspended particles in the air, which have a diameter equal to or less than 10 μm, are commonly known as PM_{10} (Kishore *et al*., 2019; Krasnov *et al.*, 2016). Being of lesser size, PM_{10} can enter the respiratory system easily and cause serious health problems like asthma, cardiovascular disease and premature deaths (Kishore *et al*., 2019; Garaga *et al.*, 2018; Ghosh *et al*., 2018; Karar and Gupta, 2006). PM_{10} can be emitted directly or indirectly through biomass burning, construction sites, soil dust, wood burning, power plants, agriculture dust, smokestacks and other processes. (Bharti *et al.*, 2017; Caseiro *et al*., 2009; Chandra *et al*., 2016; Furusjo *et al*., 2007). Over the last few decades, health-related problems have been analytically linked to deteriorating air quality due to the presence of particulate matter in the air (Jain *et al*., 2019; Marcazzan *et al.*, 2001). PM_{10} is a crucial air pollutant that deteriorates the quality of the air in metropolitan areas. (Mukherjee *et al.*, 2018; Sharma *et al.*, 2021).

According to Awasthi *et al*. (2016), the concentration of SO_2 in Haridwar has doubled and the concentration of NO_2 has increased by 2-2.5 times between 2003 and 2009. A study by Deep *et al.* (2017) concluded that the PM_{10} concentration was found to be two times higher than the national ambient air quality standards fixed by the CPCB. Kishore *et al.* (2019)

reported that NO_2 and PM_{10} had exceeded their NAAQS but SO_2 has been found within its predetermined standard in New Delhi during 2005-2012. Kotnala *et al.* (2020) discussed the lockdown impact on air pollutants in their study and found the lowest concentration of air pollutants during the lockdown periodin Delhi. A study by Srivastava *et al.* (2020) also found a significant decline in the concentration of air pollutants in Lucknow and New Delhi. The considerable drop in black carbon air pollutants in the Doon Valley, as reported by Pandey *et al*. (2022), is an indication of the role of vehicle and industrial emissions in air quality.

In recent years, Uttarakhand has experienced a significant increase in tourist activity, vehicle emissions and industrial expansion, making it imperative to study this region's air quality. The primary objective of this study is to examine the decadal behavior of trend analysis and the variability of gaseous pollutants and particulate matter (PM_{10}) along with meteorological parameters over the cities of Dehradun, Rishikesh and Haridwar in the Indian state of Uttarakhand. In this context, this study also explains the lockdown effect on gaseous pollutants and particulate matter concentrations.

2. Site description

In the present analysis, three densely populated and highly polluted major cities, namely Dehradun, Rishikesh and Haridwar in Uttarakhand, were selected as depicted in Fig. 1. Dehradun (30.32° N; 78.03° E; 700 AMSL) is the capital city of Uttarakhand state, located in the foothills of the western Himalayan region. Dehradun is a semi-industrial area, a popular tourist destination and a transit spot for millions of tourists per year visiting the Himalayas (Bhadula *et al*., 2014; Maithani *et al*., 2020). Due to more anthropogenic activities like industrialization, construction on a large scale, expeditious population growth, deforestation and unpremeditated development, air quality is getting poor in the city (Deep *et al*., 2019; Deep and Kushwaha, 2020; Kumar *et al*., 2021). For air quality monitoring, three different sites, *viz*., Clock Tower (CT), Raipur Road (RR) and Inter-State Bus Terminal (ISBT), with probably high pollution levels, were selected in Dehradun city. The CT site is the most crowded place in the city because of its location in the commercial area. The CT is located at the center of Dehradun city; hence, most vehicles pass through this site, which makes it a busy spot all the time. The RR site is a commercial and residential area that connects to a tourist spot called Maldevta and many people prefer this road to reach the airport. Hence, it is also a busy area in the city. The last site in Dehradun is ISBT, which is a commercial and industrial area. The bus depot for the city is located here, so all buses and other vehicles pass through this

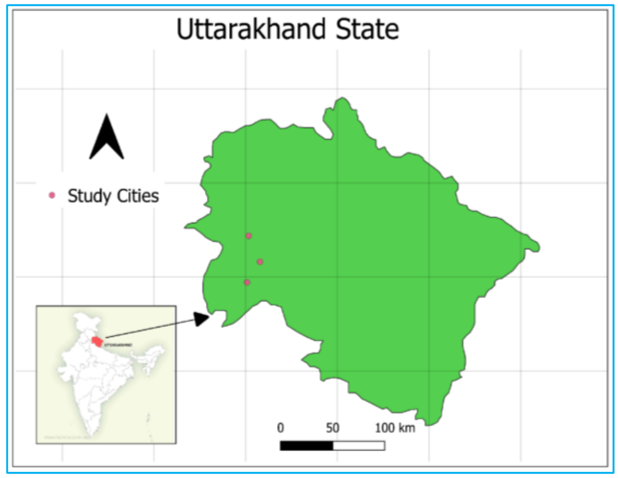


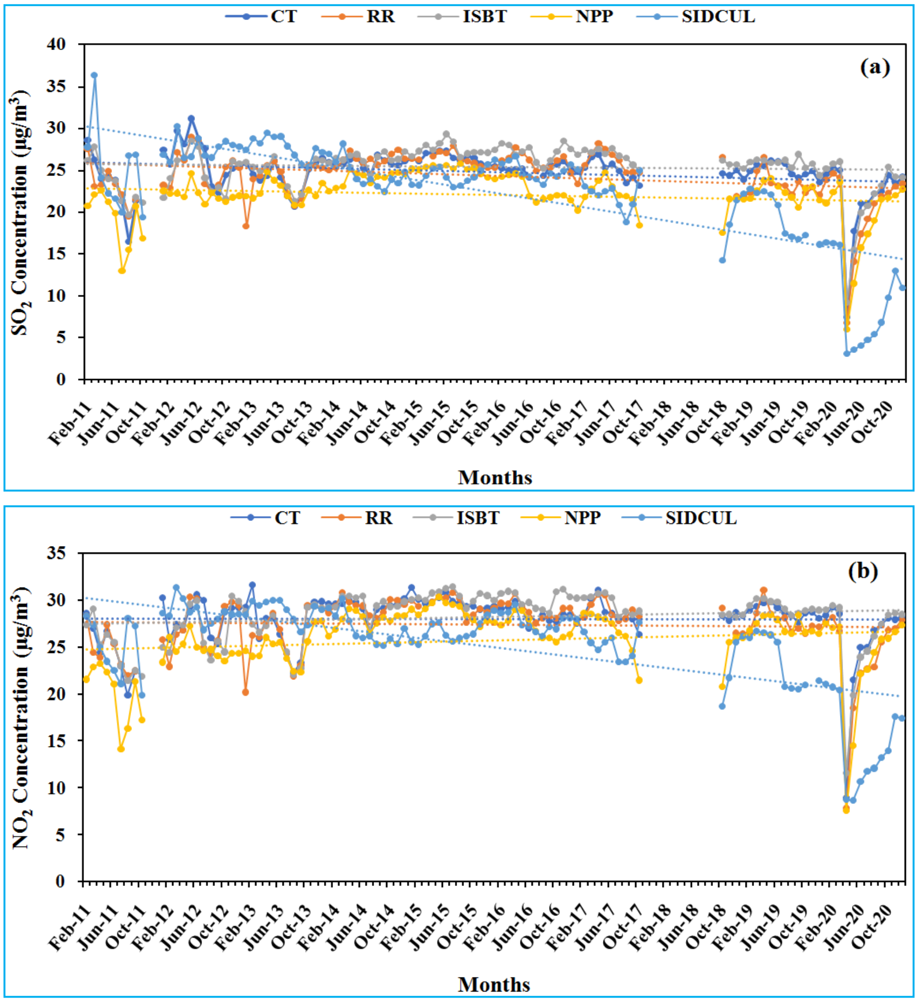
Fig. 1. Location map of Dehradun, Rishikesh and Haridwar study cities in Uttarakhand state. (India Map *Source*: www.freeworldmaps.net)

area. Due to more traffic emissions, this area is a highly polluted region of the city (Deep *et al*., 2019; Madhwal *et al*., 2020).

Rishikesh (30.09° N; 78.27° E) is located in the foothills of the Himalayas in the vicinity of the Ganga and Chandrabhaga rivers. Haridwar (29.95° N; 78.16° E; 314 AMSL); is a famous pilgrimage, near the banks of the Ganges. Nagar Palika Parishad (NPP) site was selected for the study in Rishikesh. NPP is a commercial area located near Janki Setu, which is a famous tourist spot for rafting. Across the country, people visit this place almost every season, which means more anthropogenic and traffic-related activities happen here. Haridwar is a popular destination as well as a semi-industrial area. Many people come to visit here throughout the year due to its pilgrimage sentiment. Hence, a lot of tourist and human activities happen here, which affects the air quality of this area. A SIDCUL (State Industrial Development Corporation of Uttarakhand Limited) site was selected in Haridwar City. SIDCUL is an industrial area of Haridwar City and industrial emissions directly affect the air quality of any area (Awasthi *et al*., 2016; Gautam *et al*., 2021; Chauhan *et al*., 2010).

3. Methodology and data collection

Air quality data (*viz*., concentrations of SO_2, NO_2 and PM_{10}) used in the present analysis was recorded by the Uttarakhand Pollution Control Board (http://ueppcb.uk.gov.in) during 2011-2020 every month. IMD (India Meteorological Department) in Dehradun provided daily data for meteorological parameters such as temperature, precipitation, wind speed (WS), wind direction (WD) and relative humidity (RH).



Figs. 2(a&b). Variability of the monthly mean concentration of SO_2 and NO_2 during the study period 2011-2020

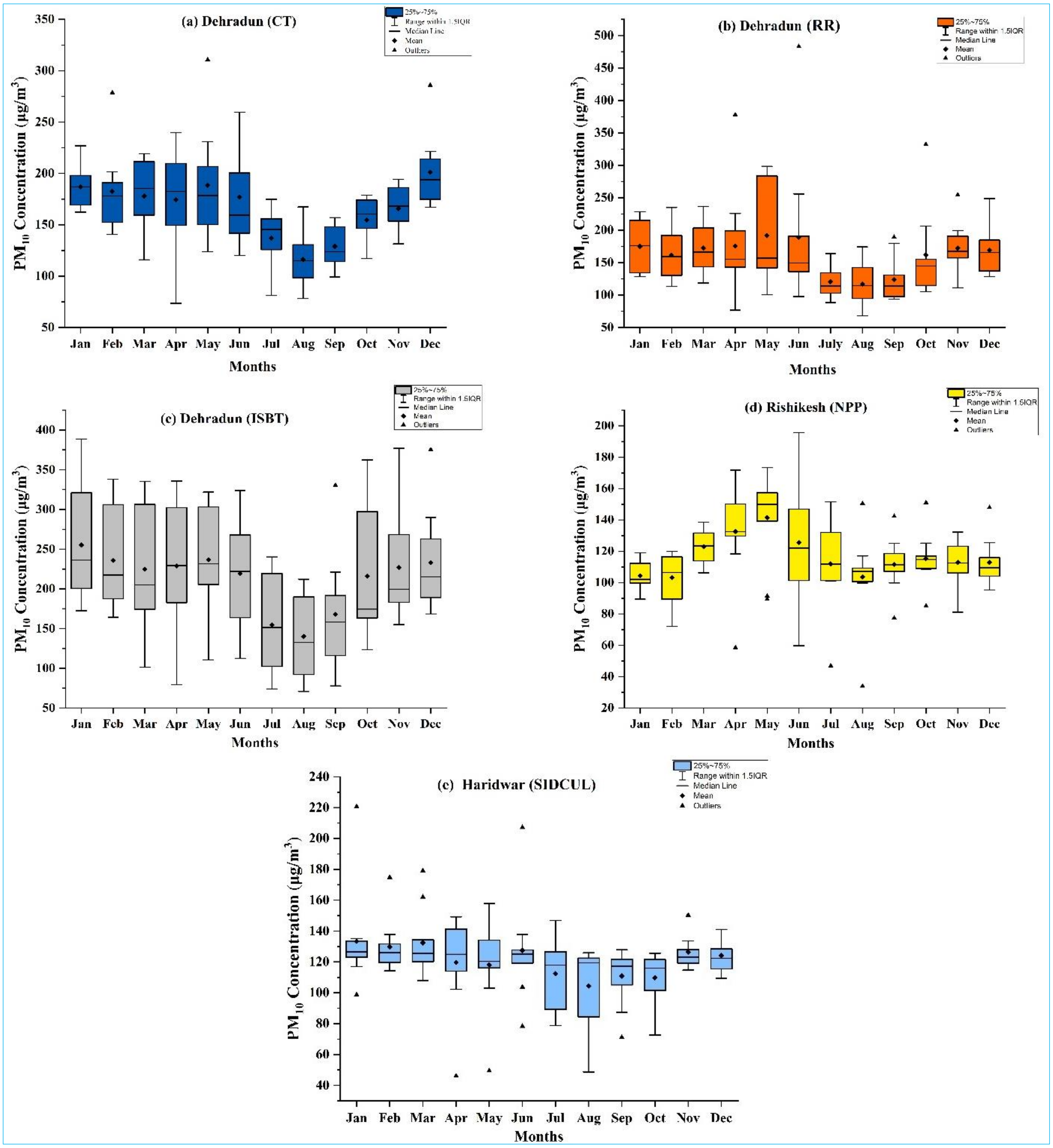
The gaseous pollutants SO_2 and NO_2 can be monitored simultaneously by a dust sampler (Joshi and Semwal, 2011). SO_2 is measured by using the modified West and Gaeke method (Chaurasia *et al*., 2020). In this method, SO_2 was absorbed in a solution of sodium tetrachloromercurate at an average flow rate of 1 per minute. The detection range of SO_2 concentration is 4-1050 µg/m^3. Nitrogen dioxide (NO_2) is measured using the Modified Jacobs and Hochheiser method (Patel *et al*., 2017), where ambient NO_2 is collected through an air-bubbling process with sodium hydroxide and sodium arsenite solution. In this method, a chemical reaction obtained by the absorption of a highly coloured dye is measured with a spectrophotometer having a range of 9-750 µg/m^3 at a length of 540 nm to ascertain the NO_2 concentration in the sample. A respirable dust sampler (Envirotech 460 NL) was used to monitor the PM_{10} concentration by the filtration-gravimetric method. The instrument was retained at a height of 2m above the ground. A Whatman filter paper (20.3 × 25.4 cm) was used in the instrument to collect the particles.

4. Results and discussion

This section has discussed the monthly, seasonal and annual variability of gaseous pollutants and particulate matter along with the variability of meteorological parameters and statistical analysis.

4.1. *Monthly and seasonal variability of gaseous pollutants and particulate matter*

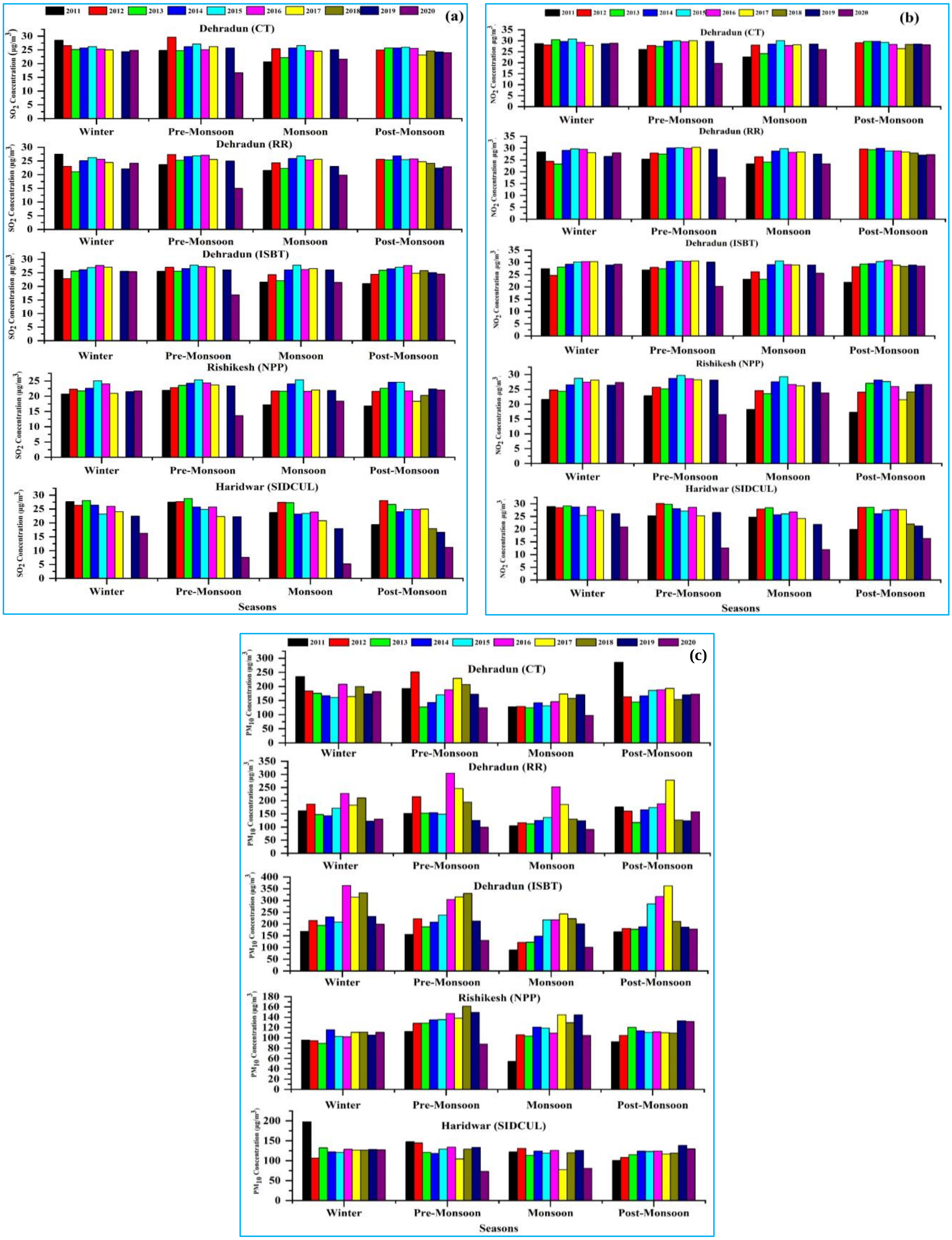
The variability of the monthly mean concentration of gaseous pollutants is depicted in Figs. 2(a&b) at selected sites. In Dehradun, the maximum concentration of SO_2 was observed in May, 2012 (31.09 µg/m^3); May, 2012 (28.88 µg/m^3); June, 2015 (29.25 µg/m^3) at CT, RR and ISBT respectively. The maximum concentration of NO_2



Figs. 3(a-e). Box-Whisker plot for the monthly mean concentration of PM_{10} during the study period 2011-2020

was observed in February, 2013 (31.66 µg/m^3); April, 2019 (31.1 µg/m^3); July, 2015 (31.42 µg/m^3) at CT, RR and ISBT sites respectively, whereas the minimum concentration was recorded in April 2020. A rapid decrement in the concentration levels of SO_2 and NO_2 was recorded in April 2020, which might be due to the COVID-19 lockdown (Venter *et al.*, 2020; Rahaman *et al.*, 2021; Sulaiman *et al.*, 2021).

In Rishikesh, the maximum concentration of SO_2 was observed to be25.73 µg/m^3 in August 2015 and the minimum concentration of SO_2 was found at 5.98 µg/m^3 in April 2020. The maximum concentration of NO_2 was found at 30.33 µg/m^3 in May, 2015 and the minimum concentration of NO_2 was observed at 7.62 µg/m^3 in April, 2020 due to lockdown conditions (Kumari and Toshniwal, 2020; Singh *et al.*, 2022).



Figs. 4(a-c). Seasonal variations of SO_2, NO_2 and PM_{10} during the study period 2011-2020

In Haridwar, the maximum concentration of SO_2 was observed to be 36.25 μg/m^3 in March, 2011 and the minimum concentration of SO_2 was found at 3.03 μg/m^3 in April, 2020. Similarly, the maximum concentration of NO_2 was found to be 31.34 μg/m^3 in March, 2012 and the minimum concentration of NO_2 was recorded at 8.68 μg/m^3 in May, 2020. During the unlocking conditions at all sites, the levels of both gaseous pollutants, SO_2 and NO_2, began to rise in June 2020.

Figs. 3(a-e) represent box-whisker plots for monthly mean PM_{10} concentrations at all study sites from 2011 to 2020. The box-whisker plot contains the mean and median as well as percentiles (25th and 75th) of the entire datasets. During the study period, the monthly mean concentration of PM_{10} ranged between 73.51 and 310.73 μg/m^3, with a mean value of 166.29 ± 42.73 μg/m^3 at CT, Dehradun. Similarly, PM_{10} was found from 67.95 to 483.18 μg/m^3 with a mean value of 160.55 ± 60.39 μg/m^3 and 70.91 to 388.67 μg/m^3 with a mean value of 211.53 ± 75.51 μg/m^3 at RR and ISBT sites respectively. The lowest PM_{10} concentration was measured at the CT site in April 2020, which could be contributed to the COVID-19 lockdown. At other sites of Dehradun, the minimum values of PM_{10} were found in August, 2012 and August, 2011 at RR and ISBT sites respectively which might be due to heavy rain in these months.

At the NPP, Rishikesh, the PM_{10} concentration was found to be between 33.98 and 195.71 μg/m^3 with a mean value of 116.62 ± 24.67 μg/m^{3}over the whole study period. Similarly, the PM_{10} concentration was found to range between 46.08 and 220.64 μg/m^3 with a mean value of 120.68 ± 24.55 μg/m^3 in SIDCUL, Haridwar. The minimum concentration of PM_{10} was recorded in April, 2020 in the SIDCUL area. Gautam *et al.* (2021) have reported that after the lockdown, the air quality of northern parts of India has improved by 30-46.67%. It is noticed that levels of PM_{10} concentration are relatively low in Rishikesh and Haridwar as compared to Dehradun city. The PM_{10} concentration may be a result of more vehicular emissions, industrial expansion and construction processes in Dehradun city (Deep *et al.*, 2019; Kishore *et al.*, 2017).

The India Meteorological Department (IMD) classifies the four seasons as winter (January to February), pre-monsoon (March to May), monsoon (June to September) and post-monsoon (October to December). Figs. 4(a-c) show seasonal variations in gaseous pollutants and particulate matter during the study period.

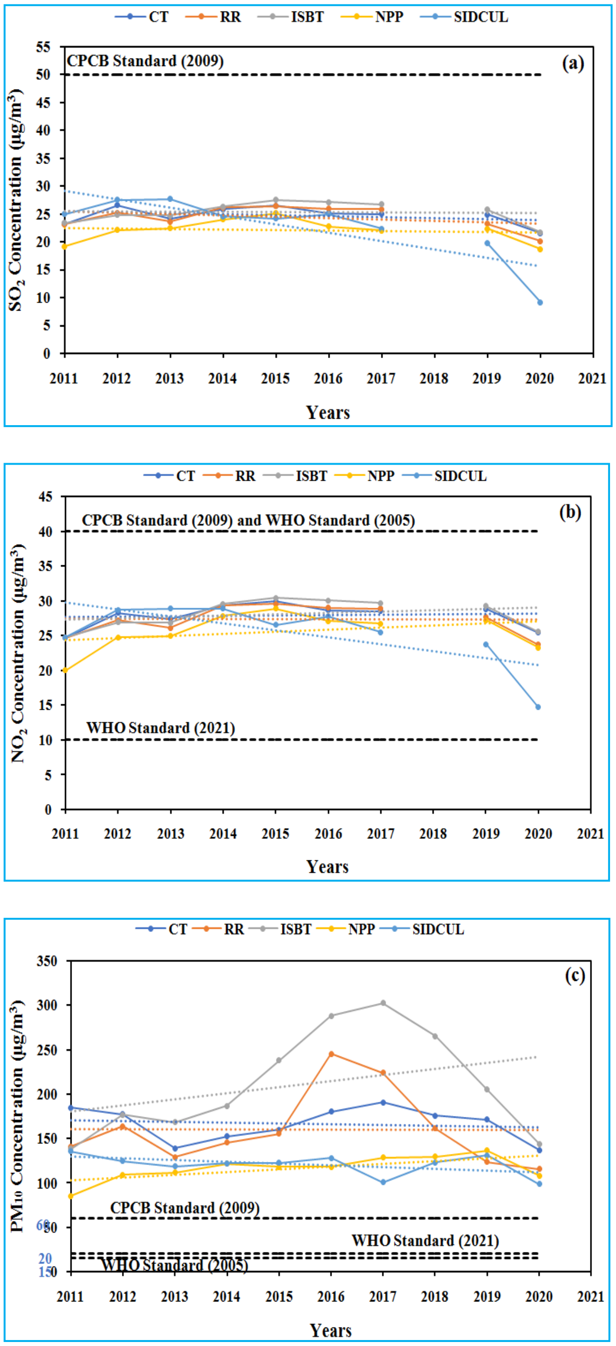
In Dehradun, the highest concentrations of SO_2 were recorded during the pre-monsoon season of 2012 (29.6 g/m^3), the winter of 2011 (27.5 g/m^3) and the pre-monsoon season of 2015 (27.83 g/m^3) at the CT, RR and ISBT sites, respectively. The lowest concentrations of NO_2 were recorded during the pre-monsoon season of 2020.

At NPP Rishikesh, the maximum seasonal concentration of SO_2 was observed to be 25.38 μg/m^3 in the pre-monsoon season of 2015 and the minimum seasonal concentration of SO_2 was found at 13.65 μg/m^3 in the pre-monsoon season of 2020. Similarly, the maximum seasonal concentration of NO_2 was found at 29.7 μg/m^3 in pre-monsoon 2015 and the minimum seasonal concentration of NO_2 was observed at 16.45 μg/m^3 in pre-monsoon 2020.

At SIDCUL Haridwar, the maximum seasonal concentration of SO_2 was observed at 28.8 μg/m^3 in the pre-monsoon season of 2013 and the minimum seasonal concentration of SO_2 was found at 5.24 μg/m^3 in the monsoon season of 2020. Similarly, the maximum seasonal concentration of NO_2 was found at 30.08 μg/m^3 in the pre-monsoon of 2012 and the minimum seasonal concentration of NO_2 was observed at 11.95 μg/m^3 in the monsoon of 2020. The concentrations of gaseous pollutants were relatively lower in Rishikesh and Haridwar as compared to Dehradun City.

The highest concentration of PM_{10} was found in the post-monsoon season of 2011(285.82 μg/m^3), pre-monsoon of 2016 (304.23 μg/m^3) and winter of 2016 (363.37 μg/m^3) at CT, RR and ISBT sites, respectively. The lowest concentration of PM_{10} was recorded during the monsoon season at all three sites of Dehradun during the study period. During monsoon season, the lowest concentration could be associated with the washout process due to the rain and local meteorological conditions (Kishore *et al.*, 2019; Ganguly *et al*., 2019; Deep *et al.*, 2018). During the post-monsoon season, crop residue burning and firecrackers are significant sources of PM_{10} (Bhuvaneshwari *et al*., 2019; Sarkar *et al*., 2018; Kishore *et al*., 2019; Kulshreshtha *et al*., 2021; Shivani *et al*., 2019).

At NPP Rishikesh, the maximum concentration of PM_{10} was observed during the pre-monsoon season of 2018 (161.24 μg/m^3) while the minimum concentration was recorded during the monsoon of 2011 (54.53 μg/m^3) during the study period. Similarly, the maximum concentration of PM_{10} was observed during the winter season of 2011 (197.74 μg/m^3) while the minimum concentration was observed during the pre-monsoon season of 2020 (72.94) at SIDCUL, Haridwar.



Figs. 5(a-c). Annual variability of SO_2, NO_2 and PM_{10} during the study period 2011-2020

4.2. *Annual variability of gaseous pollutants and particulate matter*

The annual variability of SO_2, NO_2 and PM_{10} during the study period is depicted in Figs. 5(a-c). From Fig. 5(a), we have observed that annual mean concentrations of SO_2 were below the limit of NAAQS

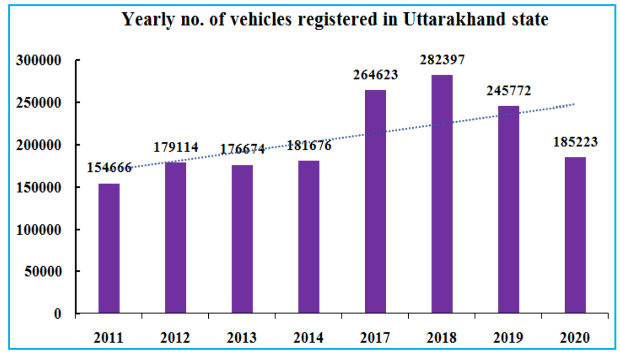


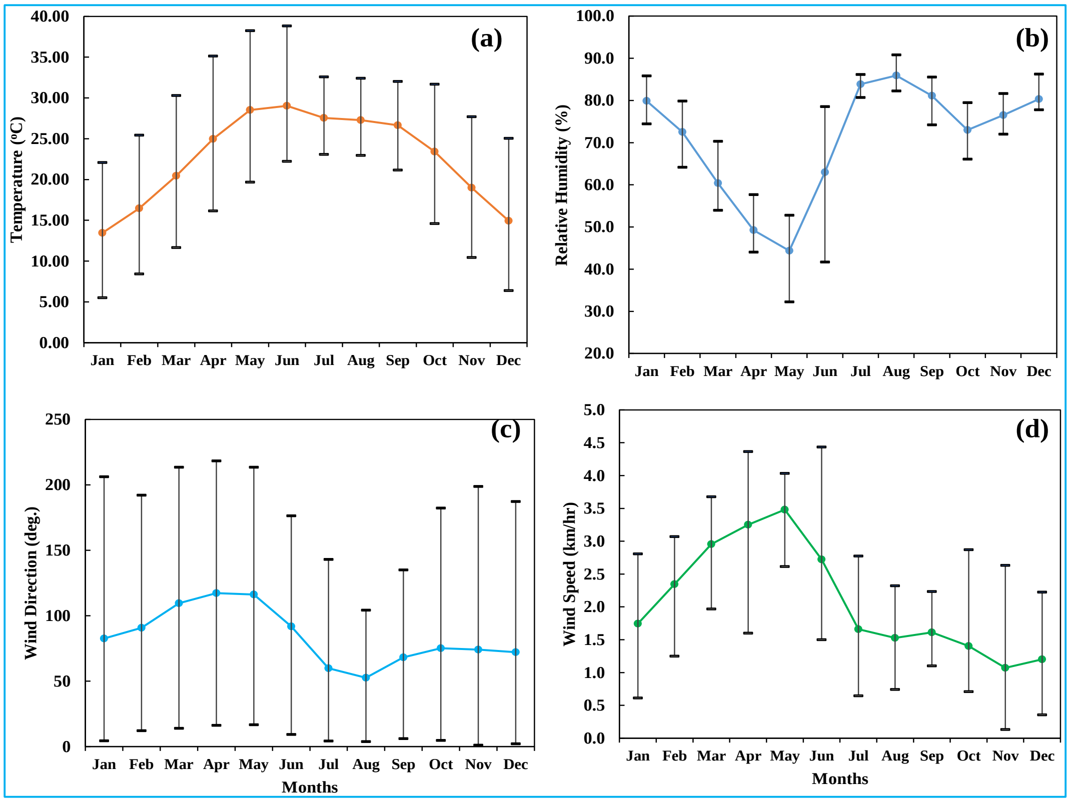
Fig. 6. Yearly number of vehicles registered in Uttarakhand state. (*Source* : State Transport Department)

(National Ambient Air Quality Standards) and the CPCB (50 µg/m^3) at all study sites, which is a good sign for the environment. From Fig. 5(b), we have concluded that levels of the annual mean concentration of NO_2 are within the NAAQS for CPCB (40 µg/m^3) and WHO standard values for 2005 (40 µg/m^3) but exceed its standard value according to WHO guidelines value 2021 (10 µg/m^3). Fig. 5(c) shows that PM_{10} concentrations are much higher than the NAAQS set by the CPCB in 2009 (60 µg/m^3), the WHO guideline value in 2005 (20 µg/m^3) and the WHO guideline for 2021 (15 µg/m^3). This is an alarming situation for human health and the environment in this region. Deep *et al.* (2019) found that PM_{10} concentration exceeded national standards during 2011-2014. Saud *et al.* (2011) have found that biomass fuels like dung cake, fuel-wood and crop residue are the major sources of PM_{10} emissions in the Indo-Gangetic Plain, India. A study by Kishore *et al.* (2017) found that the PM_{10} concentration was always higher than the NAAQ standards in Dehradun city during 2005-2012.

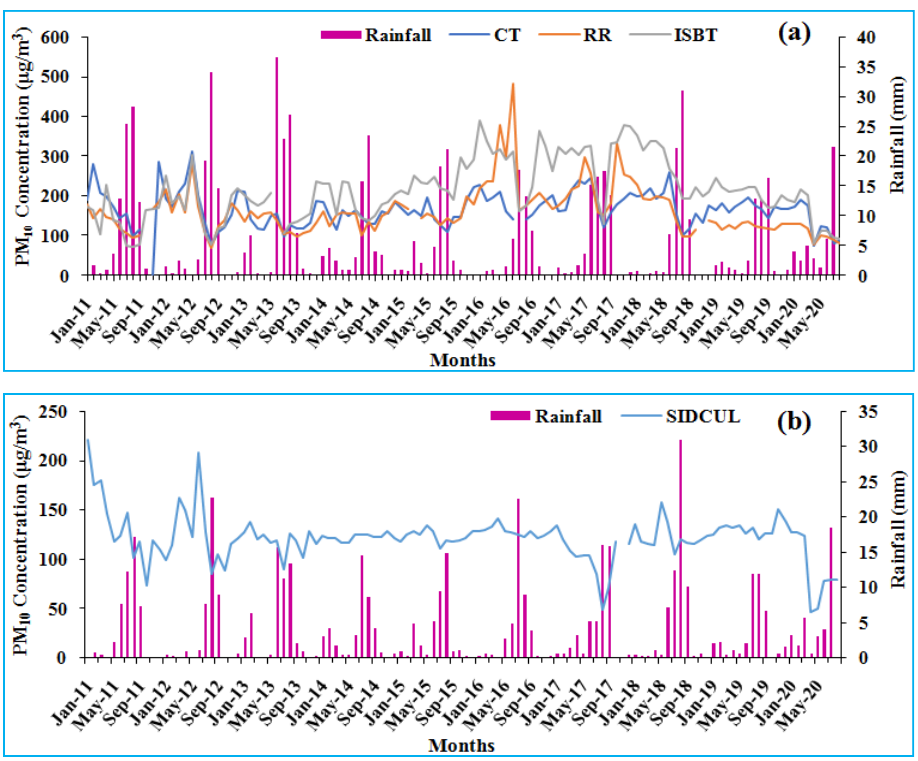
Vehicular emissions are considered a major source of air pollution in urban areas of developing countries (Ghose *et al.*, 2004; Goyal *et al.*, 2006; Biswas *et al.*, 2011). So, we have shown the total number of registered vehicles in Uttarakhand State in Fig. 6. The datasets have been obtained from the State Transport Department. From Fig. 6, the total number of registered vehicles has a positive increasing trend in Uttarakhand.

4.3. *Variability of meteorological parameters and correlation coefficients*

The mean variability of meteorological parameters of Dehradun city was studied during the study period as shown in Figs. 7(a-d). During the pre-monsoon and winter seasons, there are two peaks in the direction and speed of the wind, which could be linked to the highest levels of air pollution, especially particulate matter.



Figs. 7(a-d). Monthly mean variability of meteorology parameters Temperature, Relative Humidity, Wind speed and Wind direction at Dehradun city during the study period 2011-2020



Figs. 8(a&b). Monthly mean variations of PM_{10} with rainfall at Dehradun and Haridwar cities during the study period 2011-2020

TABLE1

The correlation coefficient between air pollutants (SO_2, NO_2 and PM_{10}) and meteorological parameters at sites CT, RR and ISBT Dehradun

Parameter	SO_2	NO_2	PM_{10}	Temp	Rainfall	RH	WS	WD
				(a) Site CT				
SO_2	1.00	**0.88**	**0.59**	-0.13	-0.30	-0.21	0.08	-0.13
NO_2		1.00	**0.42**	-0.23	-0.33	-0.08	-0.01	0.03
PM_{10}			1.00	-0.27	**-0.47**	-0.35	0.22	0.17
Temp				1.00	**0.46**	-0.30	0.31	0.06
Rainfall					1.00	**0.48**	-0.22	-0.22
RH						1.00	**-0.71**	-0.17
WS							1.00	**0.49**
WD								1.00
				(b) Site RR				
SO_2	1.00	**0.93**	**0.46**	0.03	-0.20	-0.22	0.05	-0.13
NO_2		1.00	**0.41**	-0.05	-0.27	-0.14	0.00	0.03
PM_{10}			1.00	-0.04	-0.34	-0.30	0.16	0.22
Temp				1.00	**0.46**	-0.30	0.31	0.06
Rainfall					1.00	**0.48**	-0.22	-0.22
RH						1.00	**-0.71**	-0.17
WS							1.00	**0.49**
WD								1.00
				(c) Site ISBT				
SO_2	1.00	**0.94**	**0.62**	-0.11	-0.28	-0.13	0.05	0.06
NO_2		1.00	**0.64**	-0.16	-0.34	-0.11	0.04	0.20
PM_{10}			1.00	-0.21	**-0.45**	-0.23	0.16	0.49
Temp				1.00	**0.46**	-0.30	0.31	0.06
Rainfall					1.00	**0.48**	-0.22	-0.22
RH						1.00	-0.71	-0.17
WS							1.00	**0.49**
WD								1.00

The variability of PM_{10} with rainfall in Dehradun and Haridwar are shown in Figs. 8(a&b). From Figs. 8(a&b), we observed that the lowest concentration of PM_{10} was associated with the highest rainfall. The maximum value of precipitation was recorded as 36.5 mm in June, 2013 and 30.91 mm in August, 2018 corresponding to the lowest PM_{10} concentration during these months in Dehradun and Haridwar city respectively.

To understand the effects of meteorological parameters on gaseous pollutants and particulate matter, the correlation among them has been computed, which is shown in Table 1. We have found a very strong positive correlation between the gaseous pollutants SO_2 and NO_2 (0.88, 0.93 and 0.94 at CT, RR and ISBT respectively) in Dehradun city. At CT and RR sites, a moderately positive correlation was seen between gaseous pollutants and particulate matter. A very strong positive correlation has been observed between SO_2 and PM_{10} (0.62) and between NO_2 and PM_{10} (0.64) at ISBT, Dehradun. Deep *et al.*, 2019 found a stronger correlation between gaseous pollutants SO_2 and NO_2 in Dehradun. Awasthi *et al.*, 2016

TABLE 2

Statistical analysis for air pollutants (SO_2, NO_2 and PM_{10}) during the study period 2011-2020

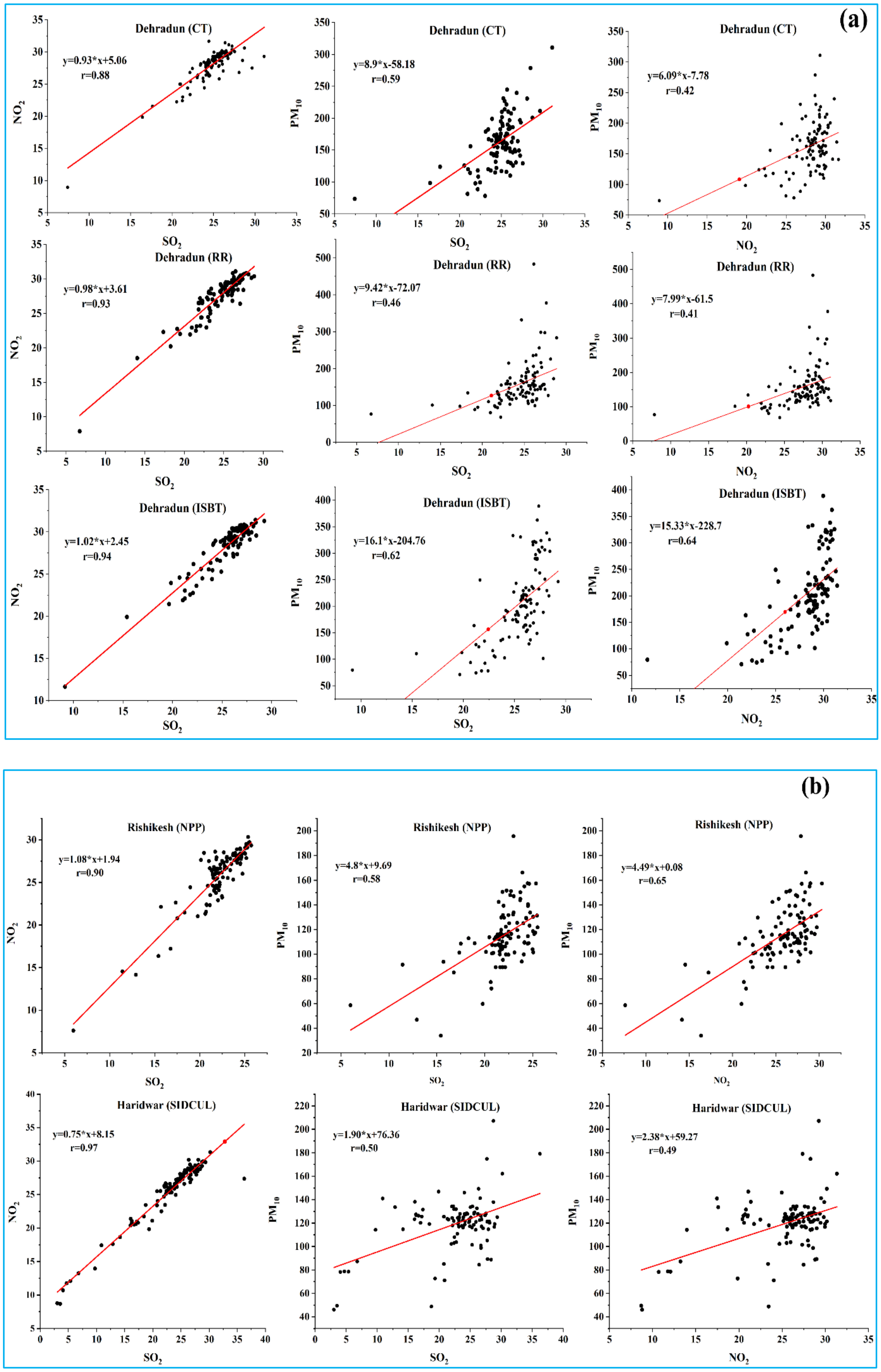
Site	Min	Max	Avg	SD	SE	Skew	Kurt
SO_2 Concentration (μg/m^3)							
CT	7.42	31.09	24.79	2.68	0.26	-2.99	17.05
RR	6.73	28.88	24.37	3.01	0.30	-2.55	11.45
ISBT	9.16	29.25	25.36	2.73	0.27	-2.76	12.04
NPP	5.98	25.73	22.06	2.90	0.28	-2.55	10.02
SIDCUL	3.03	36.25	22.59	6.06	0.59	-1.55	2.68
NO_2 Concentration (μg/m^3)							
CT	8.93	31.66	28.00	2.83	0.28	-3.52	19.36
RR	7.87	31.10	27.41	3.17	0.31	-2.81	13.24
ISBT	11.63	31.42	28.19	2.96	0.29	-2.40	8.73
NPP	7.62	30.33	25.72	3.46	0.34	-2.33	7.89
SIDCUL	8.68	31.34	25.21	4.72	0.46	-1.82	3.19
PM_{10} Concentration (μg/m^3)							
CT	73.51	310.73	166.29	42.73	3.95	0.46	0.80
RR	67.95	483.18	160.55	60.39	5.58	2.12	7.44
ISBT	70.91	388.67	211.53	75.51	6.92	0.33	-0.50
NPP	33.98	195.71	116.62	24.67	2.26	0.03	1.65
SIDCUL	46.08	220.64	120.68	24.55	2.21	0.25	4.55

have also found a strong correlation (within the range of 0.86 to 0.77) between gaseous pollutants and particulate matter at Haridwar. PM_{10} has a moderately negative correlation with rainfall (-0.47 and -0.45 at CT and ISBT sites, respectively), while a weakly negative (-0.34) relationship with rainfall has been observed at the RR site. Deep *et al.* (2017) reported that particulate matter is very strongly anti-correlated with rainfall (-0.72) at CT, Dehradun. In the case of meteorological parameters, temperature and relative humidity have a moderately positive correlation (0.46 and 0.48 respectively) with rainfall in Dehradun. There is also a very strong negative correlation (-0.71) between the relative humidity and the speed of the wind.

4.4. *Statistical analysis of gaseous pollutants and particulate matter*

For the entire study period, statistical analysis for the gaseous pollutants (SO_2 and NO_2) and particular matter (PM_{10}) is presented in Table 2. In the table, we have shown the minimum, maximum and average values of SO_2, NO_2 and PM_{10} with standard deviation (SD) and standard error (SE) during the whole study period. The SO_2 and NO_2 concentrations were found to vary from 6.73 to 31.09 μg/m^3 and 7.87 to 31.66 μg/m^3 at all sites in Dehradun during the study period. The PM_{10} concentration is observed between 67.95 and 483.18 μg/m^3 at all sites in Dehradun.

At Rishikesh, the concentrations of SO_2 and NO_2 were observed to vary from 5.98-25.73 μg/m^3 and 7.62-30.33 μg/m^3 respectively. The minimum concentration of PM_{10} was 33.98 μg/m^3 and the maximum concentration of PM_{10} was 195.71 μg/m^3. Similarly, at Haridwar, the concentration of SO_2 and NO_2 was found to vary from 3.03-36.25 μg/m^3 and 8.68-31.34 μg/m^3 respectively. The minimum concentration of PM_{10} was 46.08 μg/m^3 and the maximum concentration of PM_{10} was 220.64 μg/m^3 in Haridwar. Two other important statistical parameters for statistical calculations, such as skewness (Skew) and kurtosis (Kurt), have also been computed. Skewness measures the symmetry or asymmetry of a dataset, while kurtosis tells us about the presence of outliers in a set of



Figs. 9(a&b). Linear regression plots between gaseous pollutants and particulate matter in Dehradun, Rishikesh and Haridwar

TABLE 3

Comparison study of air pollutants (SO_2, NO_2 and PM_{10}) in various parts of India

Location	Time span	SO_2(μg/m^3)	NO_2(μg/m^3)	PM_{10}(μg/m^3)	References
Shanghai, China	2015	17	46	69	Liu *et al.*, 2020
Dhaka, Bangladesh	2016-2017	3.14 - 97.2	23.69 - 153.22	56.6 - 303	Hoque *et al.*, 2020
Split, Croatia	2017-2018	33.41	50.37	-	Bralic *et al.*, 2012
Gwalior, India	2017-2018	8.56 - 18.75	9.08 - 27.6	-	Dandotiya *et al.*, 2020
Amritsar, Punjab	2018-2019	0.62 - 59.17	1.69 - 103.38	22.33 - 896.37	Sihag *et al.*, 2022
Thapar University, Patiala	2007-2008	5 - 18	11 - 24	-	Singh *et al.*, 2010
New Delhi	2005-2012	12 ± 8	62 ± 28	254 ± 134	Kishore *et al*., 2019
Haridwar, Uttarakhand	2003-2009	4.3 - 19.35	9 - 30.32	81.92 - 402.21	Awasthi *et al.*, 2016
Dehradun, Uttarakhand	2005-2012	23.42 - 27.39	21.07 - 30.67	90.35 - 157.22	Kishore *et al*., 2017

data. Skewness can be calculated by the following formula:

$$\text{Skewness} = \frac{n\sum_{i=1}^{n}\left(X_i - \overline{X}\right)^3}{(n-1)(n-2)S^3}$$

And kurtosis is defined as;

$$\text{Kurtosis} = \frac{n(n+1)\sum_{i=1}^{n}\left(X_i - \overline{X}\right)^4}{(n-1)(n-2)(n-3)S^4} - \frac{3(n-1)^2}{(n-2)(n-3)}$$

Where X presents variables of datasets, $\overline{X}$ is the mean, n is the number of datasets and S stands for standard deviation. The skewness of datasets can be understood as follows:

Range	*Skewness of datasets*
-0.5 to 0.5	Approximately symmetric
-0.5 to -1.0 or 0.5 to 1.0	Moderately skewed
Less than -1 or greater than 1	Highly skewed

The kurtosis types can be present as follows:

Kurtosis value	*Types of kurtosis*
Kurt = 3	Mesokurtic; Excess kurtosis (0)
Kurt < 3	Platykurtic; Excess kurtosis (-ve)
Kurt > 3	Leptokurtic; Excess kurtosis (+ve)

The linear regression plots between gaseous pollutants and particulate matter have been presented in Figs. 9(a&b). In the figures, the regression equation is in the form of y = m*x + C; where m is the slope of the regression line and C represents the intercept value. For the gaseous pollutants SO_2 and NO_2, the intercept value is positive at all three sites in Dehradun. PM_{10} has negative intercept values with SO_2 and NO_2, which are observed to be extremely higher in all cases of Dehradun. At Rishikesh and Haridwar, there are all positive intercept values found between gaseous pollutants SO_2, NO_2 and particulate matter PM_{10}.

A comparison study has been shown in Table 3 to understand the levels of air pollutants at other locations. The levels of SO_2 concentration were relatively low in Shanghai (2015), Gwalior (2017-18), Thapar University, Patiala (2007-08), New Delhi (2005-12) and Haridwar (2003-09) and found to be higher in Dhaka (2016-17), Split (2017-18), Amritsar (2018-19), as compared to our study results. Similarly, the NO_2 concentration was higher than our study results except in Gwalior (2017-18) and Thapar University, Patiala (2007-08). As a comparison to our results, PM_{10} concentrations were observed lower in Shanghai (2015) and Dehradun (2005-2012).

5. Summary and conclusions

In the present study, air quality monitoring was investigated in three major cities located at the foothills of the Himalayas in the Indian state of Uttarakhand. The monthly and yearly variations of the gaseous pollutants SO_2 and NO_2 as well as the particulate matter PM_{10} in relation to changes in meteorological parameters were investigated. The COVID-19 lockdown, which reflects the direct impact of transportation, industry and human activities on gaseous pollutants, resulted in a sharp decline in pollution levels in April of 2020. Except for the year 2020, when the lowest concentration of air pollutants was recorded during the pre-monsoon season, the monsoon season saw the lowest concentration of air pollutants. The annual mean concentrations of gaseous pollutants SO_2 and NO_2 are within the CPCB-established NAAQS limits, but NO_2 concentrations at all five study sites exceeded the new WHO (2021) guideline values. During the study period, the annual mean concentration of PM_{10} exceeded the NAAQS prescribed limit at all study sites, as determined by CPCB and WHO guideline values. The higher levels of gaseous pollutants and particulate matter in the ISBT and CT sites compared to the RR could be attributed to increased vehicular emissions and human activity.

As the capital of Uttarakhand, rapid population growth, vehicular emissions, industrial expansion, increased construction activities and deforestation may be the major factors in Dehradun that directly impact the air quality of this region. We have determined that this particular particle (PM_{10}) is the most significant air pollutant in the study cities and is primarily responsible for the region's poor air quality. In the present study, it was also found that the lockdown caused by the COVID-19 pandemic contributed significantly to the improvement of the air quality in study areas. Therefore, in the future, the government should implement brief lockdowns to maintain ambient air quality. This study provides a comprehensive analysis of the current status of gaseous pollutants and particulate matter at five distinct sites in three of Uttarakhand's major cities. In the near future, this research could help policymakers control air pollution.

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Disclaimer : The contents and views expressed in this study are the views of the authors and do not necessarily reflect the views of the organizations they belong to.

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