MĀPAN-Journal of Metrology Society of India
https://doi.org/10.1007/s12647-019-00349-1

CASE REPORT

Long-Term Measurements of SO_2 Over Delhi, India

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Received: 03 May 2019 / Accepted: 13 September 2019

Abstract: Long-term measurements (2011–2018) of ambient sulphur dioxide (SO_2) and meteorology were carried out at an urban site of Delhi, India, to study the seasonal and inter-annual variations of SO_2 over Delhi. The average mixing ratio of SO_2 was estimated as 2.26 ± 0.48 ppb for the entire study period. Mixing ratio of ambient SO_2 was estimated as 2.19 ± 0.64 ppb, 2.07 ± 0.89 ppb, 2.49 ± 1.05 ppb and 2.27 ± 0.71 ppb during winter, pre-monsoon, monsoon and post-monsoon seasons, respectively. SO_2 mixing ratio was recorded maxima during monsoon (2.49 ± 1.05 ppb) season, whereas minima during pre-monsoon season (2.07 ± 0.89 ppb). The mixing ratio of SO_2 showed slightly increase in the trend during observational period. Surface wind speed and wind directions analysis indicates the influence of local sources on the mixing ratio of SO_2 at the study site. Backward trajectories and potential source contributing factor (PSCF) analysis also showed the local as well as the regional sources (industrial activities, coal burning and thermal power plants etc.,) influencing the mixing ratio of SO_2 over Delhi.

Keywords: Ambient SO_2; Seasonal variation; Trajectory analysis; PSCF analysis

1. Introduction

Sulphur dioxide (SO_2) is a primary criterion air pollutant and a major precursor of sulphate aerosols that plays an important role in tropospheric chemistry and climate change. SO_2 also plays a vital role in earth's radiative balance (IPCC 2013), which is responsible for the local air pollution and a contributor to acid rain as well as smog formation [1]. The major anthropogenic source of SO_2 is combustion of coal in thermal power plants, smelters, pulp mills, petroleum refineries and industries [2]. SO_2 and sulphate particles (SO_4^{2-}) are more harmful when inhaled together [3]. Acid rain which occurs as a result of photochemical oxidation of sulphur and nitrous oxide changes the chemical balance of the soil and damages the agricultural crops and forest [4]. SO_4^{2-} particles can contribute to a reduction in visibility and discoloration of the atmosphere; however, majority of the damage from particle pollution comes from acid deposition. The wet deposition of acidic SO_4^{2-} particles results in the acidification of surface water and subsequent damage to aquatic ecosystems, forests, vegetation and building materials and structures [5].

Mixing ratio of SO_2 is observed to decrease in some part urban area of the India, South East Asia, China and Africa; however, the anthropogenic SO_2 emissions are poorly controlled and characterized [6]. Different urban regions of India show large variation in mixing ratio of ambient SO_2. Due to its significant climatic impacts and sulphate aerosols formation over the region, mixing ratio of ambient SO_2 is poorly characterized over Delhi on long-term basis. In the present paper, we report the diurnal, seasonal and annual variations in mixing ratio of SO_2 at an urban site of Delhi, India, from January 2011 to December 2018. In addition, we report the influence of meteorology on mixing ratio of SO_2 over Delhi. The HYSPLIT model is also used to explore the possible potential source regions of SO_2 to the measurement site of Delhi.

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s12647-019-00349-1) contains supplementary material, which is available to authorized users.

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Published online: 05 November 2019

2. Methodology

2.1. Site Description

SO_2 was measured at CSIR-National Physical Laboratory (28°38′N, 77°10′E; 218 m amsl), New Delhi, India, from January 2011 to December 2018 (Fig. 1). Delhi, the capital of India, is located (28.61°N, 77.23°E) in the northern part of the country. Vehicular emissions, industrial emissions, secondary aerosols, biomass burning, etc., are considered to be major sources of pollutants over Delhi [7]. During 2017–2018, the total number of vehicles registered in Delhi was ~ 11 million [8]. Generally, Delhi experiences four major seasons: winter (January–February), pre-monsoon (March–May), monsoon (June–September) and post-monsoon (October–December). A brief summary of meteorological parameters (temperature, relative humidity, wind speed and wind directions) of measurement site is given in Table 1. During winter (average temperature: 14.6 ± 2.5 °C), this area is influenced by winds from NE to NW and in pre-monsoon (average temperature: 28.8 ± 2.7 °C) from SE to SW. A detailed description of measurement site is available in Sharma et al. [9–11].

2.2. Experimental Setup

The measurements of SO_2 and meteorological parameters (temperature, relative humidity (RH), wind direction and wind speed) were carried out using SO_2 analyser and automatic weather station (AWS), respectively, from January 2011 to December 2018. Ambient SO_2 was measured using calibrated SO_2 Analyser (Model: APSA 360A, M/s. Horiba Ltd, Japan) working on ultraviolet fluorescence method [6]. The instrument was calibrated automatically every day using the in-built calibrator for zero and span (in-built permeation tube/oven). The lower detection limit (LDL) of the instrument was ± 0.5 ppb at a flow rate of 0.8 l min^{-1}. The calibration was also performed periodically with zero gas and span gas using NIST traceable certified SO_2 gas (500 ppb ± 5%). Zero Air Generator

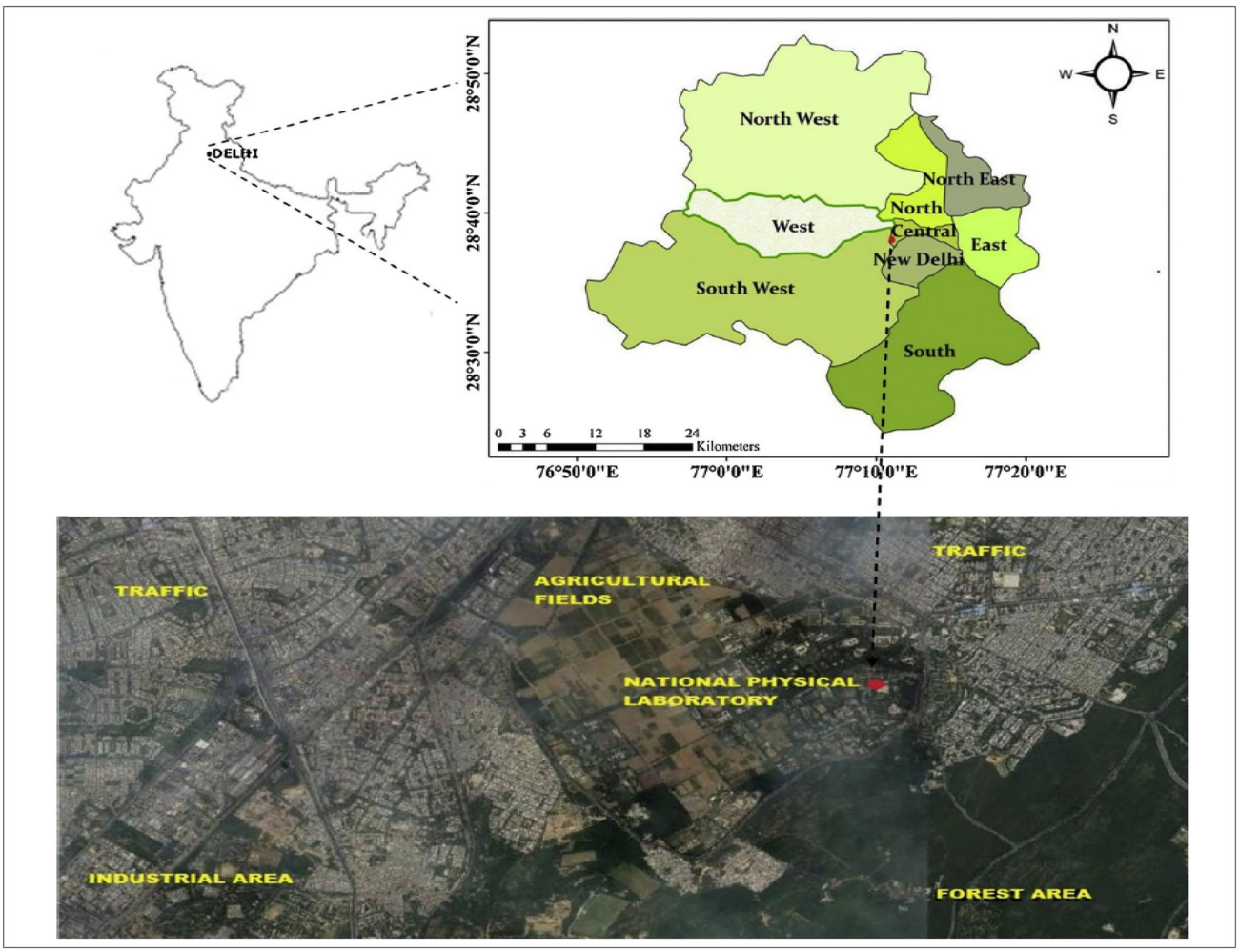

Fig. 1 Map of observational site

Table 1 Seasonal variation of mixing ratio of SO_2 and its range along with meteorological parameters (temperature, RH and wind speed) during 2011–2018

Parameters	Winter	Pre-monsoon	Monsoon	Post-monsoon
SO_2 (ppb)	2.19[b] ± 0.64 (1.22–5.25)	2.07[b] ± 0.89 (0.82–4.14)	2.49[a] ± 1.05 (1.22–5.06)	2.27[b] ± 0.71 (1.11–3.83)
Temp (°C)	14.6 ± 2.4 (7.2–24.01)	28.8 ± 2.7 (15.5–42.8)	30.5 ± 2.0 (24.2–40.1)	22.4 ± 3.25 (10.4–31.9)
RH (%)	68.8 ± 12.4 (35.7–88.1)	36.1 ± 11.1 (12.7–75.7)	60.6 ± 12.0 (20.5–85.8)	51.6 ± 10.63 (23.6–77.2)
WS (m s^{-1})	1.34 ± 0.61 (0.6–2.3)	1.73 ± 0.85 (0.6–2.78)	1.77 ± 0.81 (0.7–3.41)	1.21 ± 0.59 (0.31–2.15)

± Standard deviation

[a]Significantly different ($p < 0.05$)

[b]Not significantly different ($p > 0.05$)

(Model: PAG-003, M/s. ECO Physics AG, Switzerland, accuracy ± 0.01 ppb) has been used for zero air calibration. The details of the operating, calibration and maintenance of the SO_2 analyser are discussed in our previous publication [6]. The instrument was operated continuously (24 h) with an uninterrupted steady power supply for more than 15–20 days in a month (1 week time used for general maintenance, routine check and calibration). Statistical analysis was carried out to test the annual and seasonal variability of SO_2 using Chi-square method by Monte Carlo statistics as well as ANOVA method.

In order to identify the possible pathway of the air parcel to estimate the long-range transport of pollutants to the measurement site, backward trajectories are computed. In the present case, 72-h backward air mass trajectories were calculated using the HYSPLIT (HYbrid Single Particle Lagrangian Trajectory) model [12, 13]. Trajectories were computed from January 2011 to December 2018 starting at 1000 h Indian Standard Time (IST) at 500 m, above the ground level (AGL). This height was chosen to diminish the effects of surface friction and to represent winds in the low boundary layer. Trace gases (SO_2) have the ability to travel long distance; therefore, 3 days were selected to calculate backward trajectories [14]. The detailed methods of trajectories and PSCF analysis are available in our previous publication and reference therein [6, 9–11].

3. Results and Discussion

3.1. Mixing Ratio of SO_2

Average mixing ratio of SO_2 was 2.26 ± 0.48 ppb with a range of 0.82–5.25 ppb for the entire study period. Annual average mixing ratio of SO_2 was 2.41 ± 0.36 ppb, 2.23 ± 0.43 ppb, 1.87 ± 0.36 ppb, 2.29 ± 0.28 ppb, 2.34 ± 0.52 ppb, 2.19 ± 0.75 ppb, 2.31 ± 0.71 ppb and 2.46 ± 0.68 ppb during 2011, 2012, 2013, 2014, 2015, 2016, 2017 and 2018, respectively (Table 2). The annual average mixing ratio as well as the inter-annual variation (annual variation with respect to average value of entire measurement period) with statistics of SO_2 is summarized in Table 2. The highest annual average mixing ratio of SO_2 was 2.46 ± 0.68 ppb in 2018, whereas minimum annual average mixing ratio of SO_2 was 1.87 ± 0.36 ppb in 2013. In the present study, the non-significant inter-annual variation in mixing ratio of SO_2 was recorded at measurement site of Delhi except 2013 and 2018 (Table 2). Figure 2

Table 2 Annual average as well as inter-annual differences with average mixing ratio of SO_2 from 2011 to 2018

Year	SO_2 (ppb)	Inter-annual difference (ppb)	Changes in mixing ratio of SO_2 (%)
2011	2.41 ± 0.36	0.15[b]	6.6
2012	2.23 ± 0.43	− 0.03[b]	1.3
2013	1.87 ± 0.36	− 0.39[a]	17.2
2014	2.29 ± 0.28	0.03[b]	1.3
2015	2.34 ± 0.52	0.08[a]	3.5
2016	2.19 ± 0.75	0.07[b]	3.1
2017	2.31 ± 0.71	0.05[b]	2.2
2018	2.46 ± 0.68	0.20[a]	8.9
Average	2.26 ± 0.48	0	0

± Standard deviation

[a]Significantly different ($p < 0.05$)

[b]Not significantly different ($p > 0.05$)

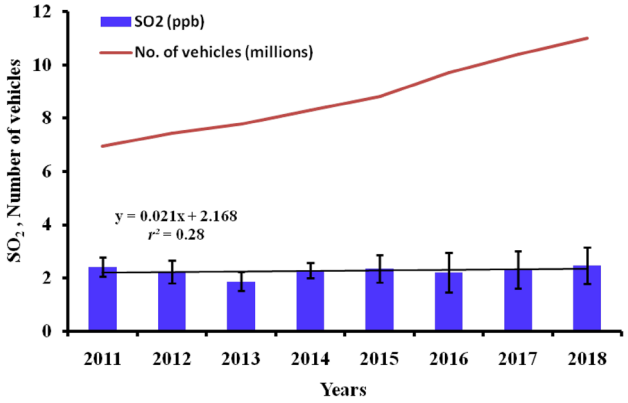

Fig. 2 Trend of annual SO_2 mixing ratio and total number of registered vehicles in Delhi from 2011 to 2018

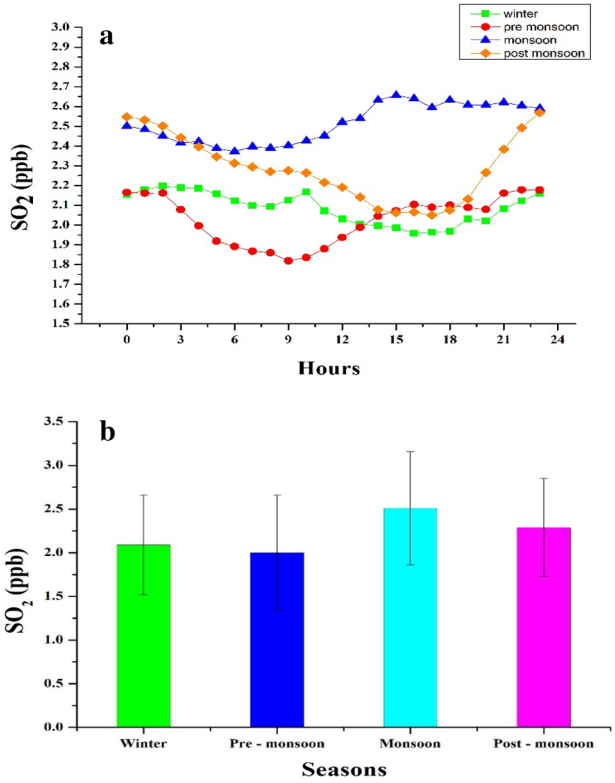

Fig. 3 **a** Diurnal and **b** seasonal variations in mixing ratio of SO_2 in Delhi

shows slight increase in trend of SO_2 mixing ratio in comparison to the total number of registered vehicles in Delhi from 2011 to 2018. The total numbers of registered vehicles that spiked up to almost twice (2011 to 2018) in Delhi indicates the minimum influence of vehicular emissions on mixing ratio of SO_2 during the study period. The inter-annual variation in mixing ratio of SO_2 was might be due to source strength of SO_2 and meteorological condition of the measurement site. However, the mixing ratio of SO_2 was recorded to be well within the permissible limit of National Ambient Air Quality Standards (NAAQS) of India (annual: 50 μg m^{-3} or 20 ppb) during the entire observational period. Datta et al. [6] reported the mixing ratio of ambient SO_2 as 1.76 ± 0.56 ppb (in 2008), whereas Saraswati et al. [15] reported as 1.74 ± 0.53 ppb (in 2013–15) over Delhi. The similar results have been reported in other studies at different sites of Delhi [4, 16–21]. Chelani and Devotta [17] reported the average mixing ratio of SO_2 as 7.5 ppb (average of 10 observational site) during 2000–2003, whereas Khaiwal et al. [20] reported the mixing ratio of SO_2 as 5–7 ppb during 1998–2003 over Delhi region (before implementation of CNG). The reduction in mixing ratio of SO_2 in Delhi after 2001 may be due to conversion of diesel fuel in public vehicles to CNG, rapid transit system—Delhi Metro–and adoption and other policies [22]. Badami [23] reported that the mixing ratio of SO_2 decreased by 33% after implementation of CNG fuel and reduction in the sulphur content in diesel.

The night-time mixing ratio of SO_2 was observed to be high across all the seasons (Fig. 3a). The daytime average mixing ratio of SO_2 was recorded as 2.04 ± 0.58 ppb, 1.95 ± 0.66 ppb, 2.50 ± 0.71 ppb and 2.19 ± 0.55 ppb during winter, pre-monsoon, monsoon and post-monsoon, respectively. The night-time average mixing ratio of ambient SO_2 was recorded as 2.12 ± 0.56 ppb, 2.11 ± 0.67 ppb, 2.53 ± 0.58 ppb and 2.39 ± 0.57 ppb, for winter, pre-monsoon, monsoon and post-monsoon seasons, respectively. The higher SO_2 mixing ratio during night-time can be attributed to the significant reduction in the atmospheric boundary layer mixing height (in winter: ∼ 100 m; pre-monsoon: ∼ 250 m; monsoon: 400 m and post–monsoon: 250 m) as well as source strength of SO_2 at measurement site of Delhi.

The average mixing ratio of SO_2 was recorded as 2.19 ± 0.64 ppb, 2.07 ± 0.89 ppb, 2.49 ± 1.05 ppb and 2.27 ± 0.71 ppb during winter, pre-monsoon, monsoon and post-monsoon seasons, respectively (Table 1 and Fig. 3b). The seasonal variation in mixing ratio of ambient SO_2 and meteorological parameters (temperature, RH, wind speed and wind direction) with their ranges are summarized in Table 1. In the present case, non-significant seasonal variation of SO_2 was observed during all the seasons except monsoon season (Table 1). The seasonal changes in mixing ratio of SO_2 may be due to source strength and prevailing meteorological conditions at the observational site [6, 7]. The higher mixing ratio of ambient SO_2 was recorded during monsoon season followed by post-monsoon, winter and pre-monsoon seasons.

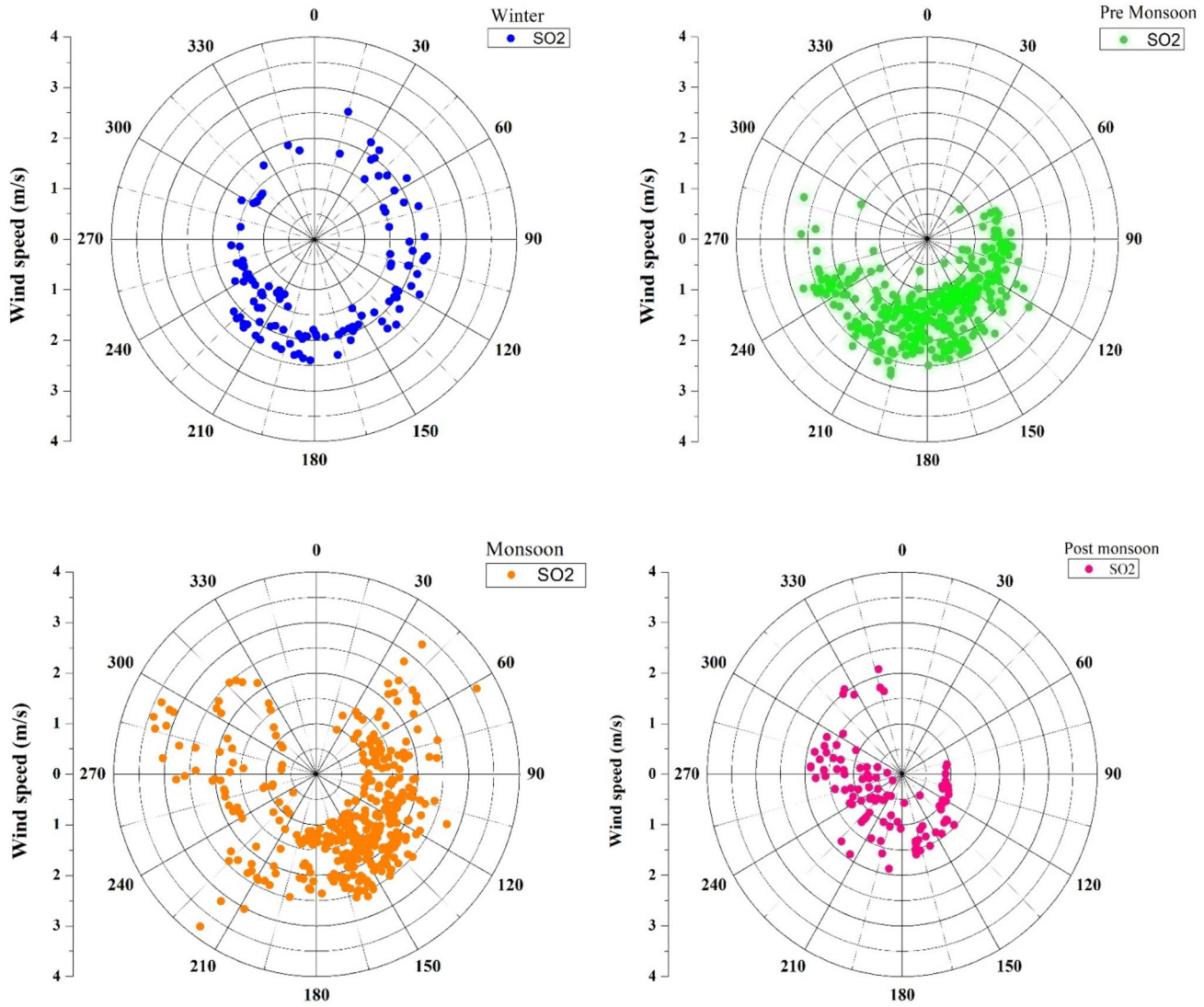

Fig. 4 Bivariate polar plots between wind direction and wind speed during winter, pre-monsoon, monsoon and post-monsoon seasons

3.2. Effect of Meteorology on SO_2

The variations in the local meteorological conditions (temperature, RH, wind direction and wind speed) may influence the variation of ambient SO_2 at the observational site of Delhi. Therefore, analysis of meteorological parameters in measured species is very helpful for the better understanding of local and regional occurrence/source of SO_2. The seasonal and long-term summary of meteorological parameters measured from January 2011 to December 2018 is summarized in Table 1. During all the seasons, mixing ratio of ambient SO_2 is non-significantly negatively correlated with ambient temperature (Table S1, in supplementary information). The wind is one of the most important parameters for the transfer and dispersion of pollutants. In the present study, ambient SO_2 was non-significantly negatively correlated with wind speed during all the seasons. The distribution of average surface wind speed with the wind direction, during winter, pre-monsoon, monsoon and post-monsoon seasons, is depicted in Fig. S1 (in supplementary information). Negative correlation between wind speed and SO_2 mixing ratio is an indicator of local source domination [24] and dilution of SO_2 by the wind [25, 26].

3.3. Possible Source Region of SO_2

To explore the possible sources and source region of ambient SO_2 at the observational site of Delhi, surface wind analysis, wind direction, trajectories, cluster, and PSCF analysis have been performed.

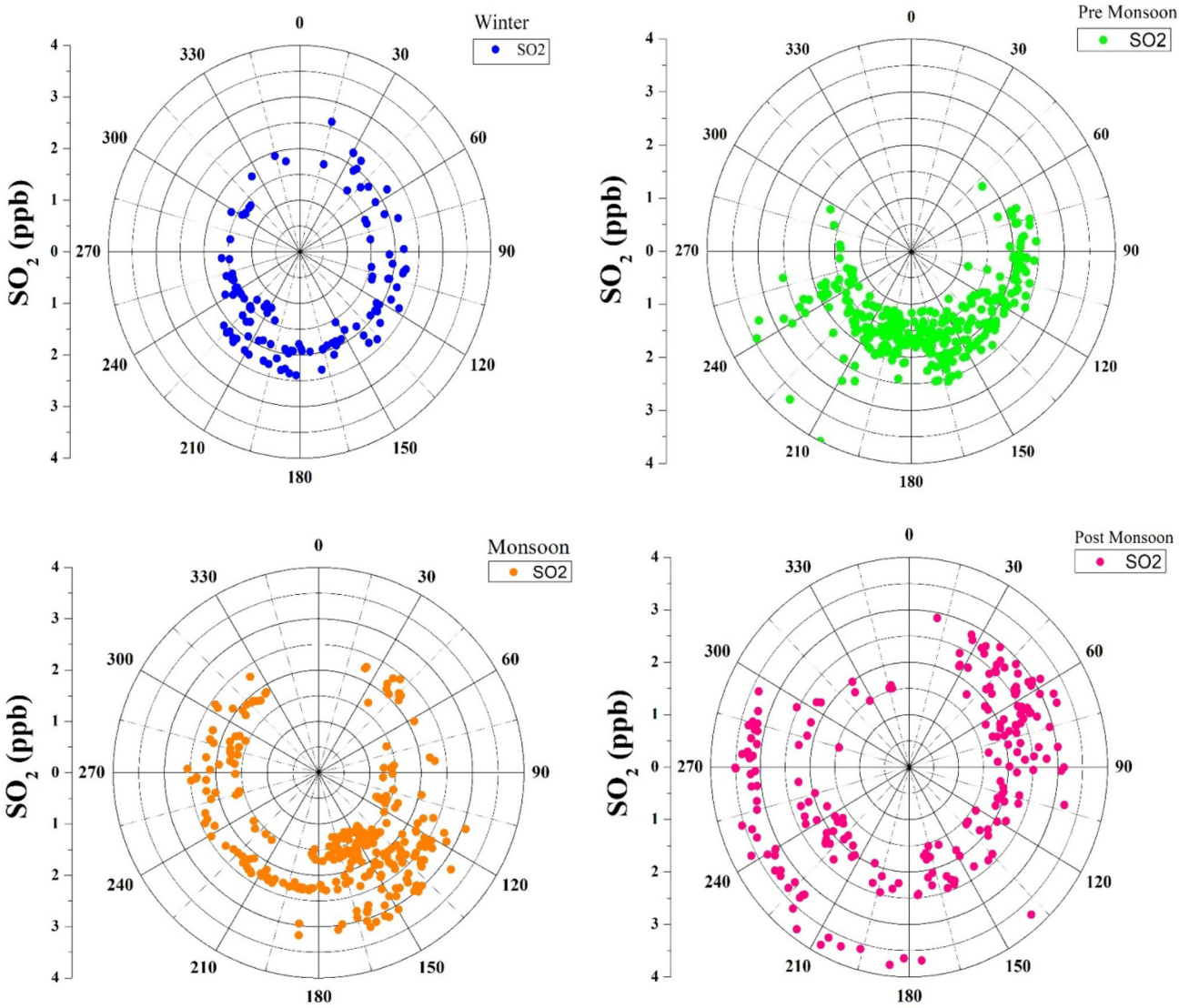

Fig. 5 Bivariate polar plots between wind direction and SO_2 during winter, pre-monsoon, monsoon and post-monsoon seasons

The highest SO_2 mixing ratio was observed during the lowest wind speed (1–2 m s^{-1}) from downwind direction which indicates the possible nearby sources (from NE, S and SW directions as well as from NW direction) during all the seasons (Figs. 4, 5). The highest SO_2 mixing ratio was observed during monsoon season may be due to the favourable higher wind conditions (from NE, W, SW and NW directions) as well as regional contributions (from coal burning, industries, power plants, etc.). Coal-based thermal power plants (TPP) in Delhi-NCR are summarized in Table 3 with direction towards the observational site. The emissions from the coal-fired thermal powers (from NW, SW and SE directions) over the Delhi-NCR region influenced the mixing ratio of SO_2 in the observational site of Delhi. In order to identify the possible source regions and pathway of SO_2 in the observational site of Delhi, 72-h backward trajectories were calculated using HYSPLIT model [12] as depicted in Fig. 6. The trajectory analysis indicates that the air mass approaching the observational site during entire sampling period is mainly from Indo-Gangetic plain (IGP) region (Uttar Pradesh, Haryana and Punjab), Thar Desert, Afghanistan, Pakistan and surrounding areas (Figs. 6, 7). During winter season the approaching air mass at the receptor site is mainly of continental type and transported from the IGP, Pakistan, Afghanistan and its surrounding areas (Fig. 6). Sharma et al. [27] also observed the similar trajectories at Delhi during winter 2012. During pre-monsoon, the approaching air mass at the receptor site is mainly from Rajasthan (Thar Desert), Gujarat, Pakistan and Arabian Sea, whereas during monsoon season, it is approaching from IGP, Rajasthan, Arabian Sea, Bay of Bengal and surrounding areas (cluster

Table 3 Coal-based thermal power plants (TPP) in Delhi and its surroundings

ID	Name and locations	Distance (km)	Directions	Status
1.	Badarpur Thermal Power Plant, Delhi	20	SW	Closed in 2018
2.	Rajghat Thermal Power Plant, Delhi	8	E	Closed in 2015
3.	Panipat Thermal Power Plant, Panipat	89	NW	Operational
4.	National Thermal Power Station, Dadri	42	SE	Operational
5.	Aravali Thermal Power Plant, Jhajjar	80	SW	Operational
6.	Mahatma Gandhi Thermal Power Station, Jhajjar	81	SW	Operational

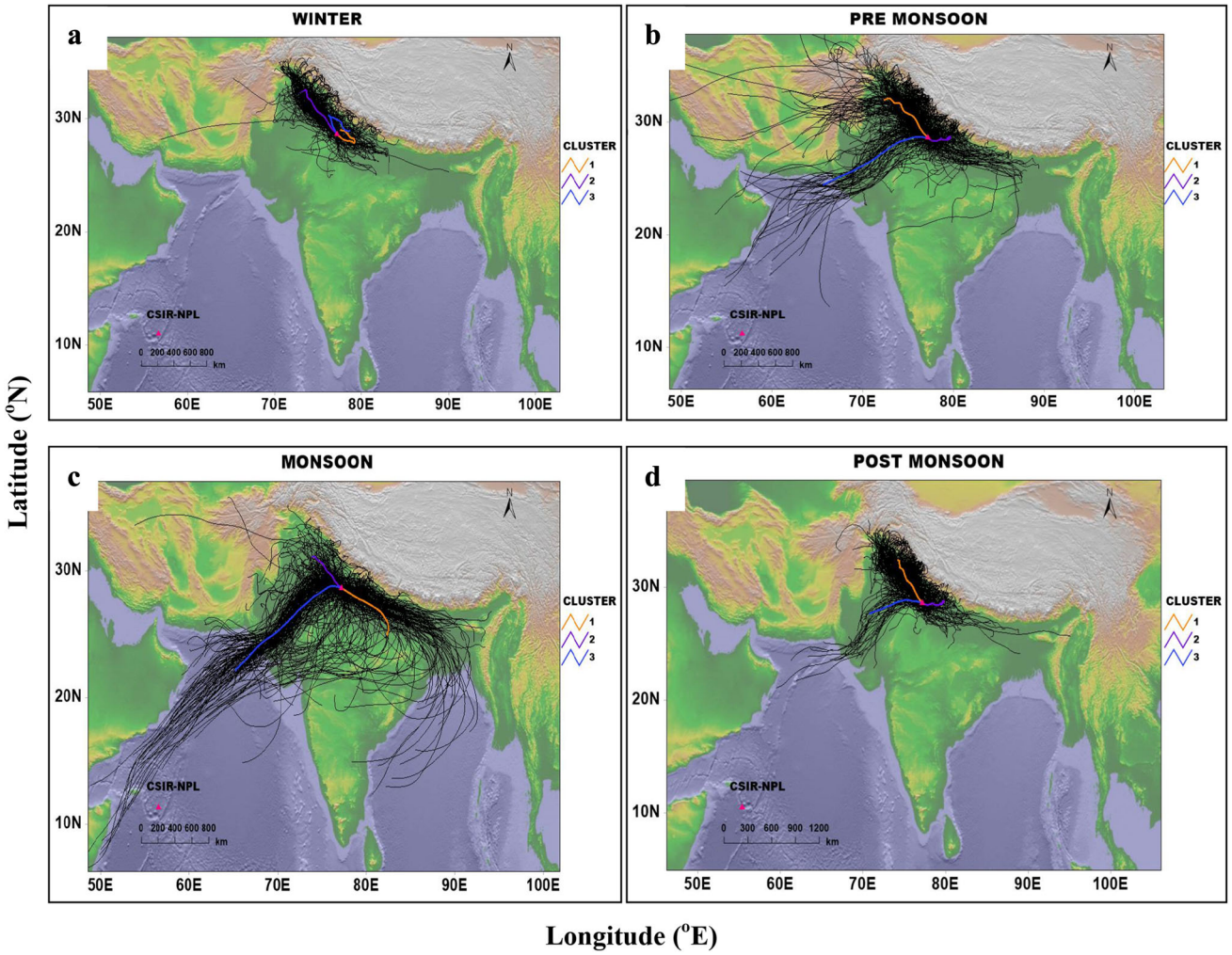

Fig. 6 Three-day backward trajectory and cluster analysis of air mass during **a** winter, **b** pre-monsoon, **c** monsoon and **d** post-monsoon seasons

analysis: Table S2, in supplementary information). During monsoon season, winds with the combined effect of continental and marine air mass has flown to the observational site of Delhi. In all the cases, the air mass backward trajectories support the observations for their origin. Cluster analysis as well as PSCF analysis indicates the same source region (Figs. 6, 7). Furthermore, the pollutants from the burning of biomass in these regions are transported to the observational site [28]. This is evident from the post-monsoon PSCF analysis which indicates higher contribution from the states of Punjab and Haryana and can be supported by the biomass burning occurring during October–November.

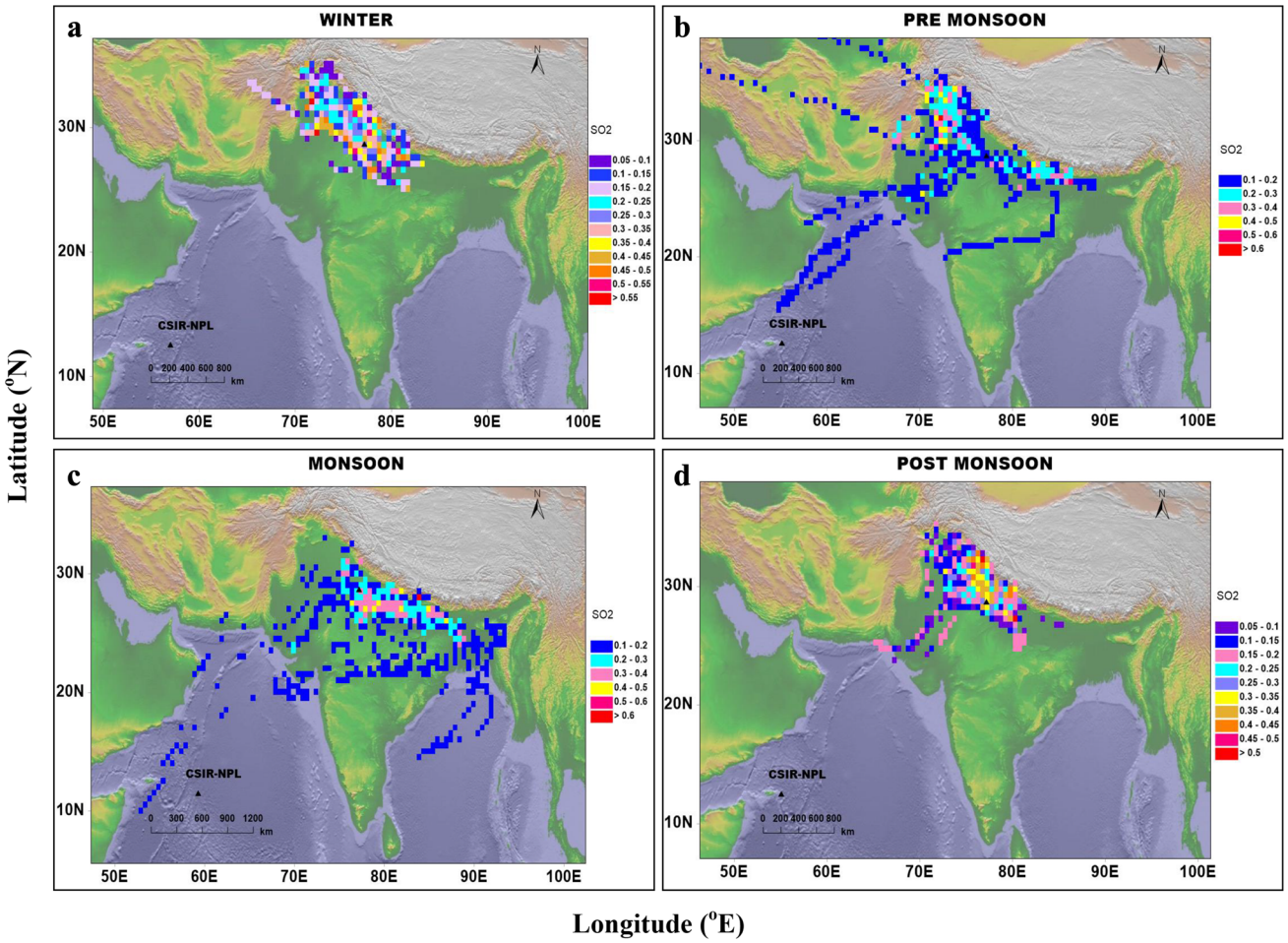

Fig. 7 PSCF analysis of SO_2 during winter, pre-monsoon, monsoon and post-monsoon seasons in Delhi

4. Conclusions

In the present study, the average mixing ratio of SO_2 was 2.26 ± 0.48 ppb for the entire study period. The non-significant diurnal, seasonal and inter-annual variation in the mixing ratio of SO_2 was observed at Delhi. Analysis of surface wind speed and wind directions with SO_2 indicates that the sources of nearby areas (industrial activities, coal burning, thermal power plants, etc.) contributed significantly to SO_2 mixing ratio, whereas trajectories and PSCF analysis showed that the regional sources along with local sources influenced the SO_2 mixing ratio at the observational site of Delhi. The contribution of SO_2 was also from the highly urbanized and populated regions of NCR of Delhi, Punjab, Haryana and Uttar Pradesh, probably due to many thermal power plants located over these regions. In addition, the transported air mass from the burning of agricultural crop residues in the associated areas may have influenced the mixing ratio of SO_2 at the observational site of Delhi. Although in this study, the SO_2 mixing ratio do not breach the NAAQS guidelines whereas, it might become a concern because of the increasing rate of sulphate aerosols as well as haze formation over the region.

Acknowledgements The authors are thankful to the Director, CSIR-National Physical Laboratory, New Delhi, and Head, Environmental Sciences and Biomedical Metrology Division, CSIR-NPL, New Delhi, for their encouragement and support for this study. The authors thankfully acknowledge the NOAA for downloading the trajectories datasets (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1) and HYSPLIT model. The authors are thankful to the anonymous reviewers for their constructive suggestions to improve the manuscript.

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