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Influence of meteorology and interrelationship with greenhouse gases (CO₂ and CH₄) at a sub-urban site of India

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due to high uncertainty in its sources and sinks (Keppler et al., 2006; Miller et al., 2007; Frankenberg et al., 2008). Kirschke et al. (2013) reported that in India, agriculture and waste constitutes the single largest regional source of CH_4 . Although many sources and sinks have been identified for CH_4 , their relative contribution to atmospheric CH_4 is still uncertain (Garg et al., 2001; Kirschke et al., 2013). In India, electric power generation that contributes to half of India's total CO_2 equivalent emissions (Garg et al., 2001).

Global climate change has serious impact on humans and ecosystems. Due to this, many factors have been identified that may reflect or cause variations in environmental change (Pielke et al., 2002). Out of these, the Normalized Difference Vegetation Index (NDVI) has become one of the most widely used indices to represent the biosphere influence on global change (Yang et al., 2011). The planetary boundary layer (PBL) is the part of the atmosphere closest to the Earth's surface where turbulent processes often dominate the vertical redistribution of sensible heat, moisture, momentum, and aerosols/pollution (Ao et al., 2012).

Greenhouse and other trace gases have great importance in atmospheric chemistry and for radiation budget of the atmosphere–biosphere system (Crutzen et al., 1991). Hydroxyl radicals (OH) are very reactive oxidizing agents, which are responsible for the oxidation of almost all gases that are emitted by natural and anthropogenic activities in the atmosphere. Atmospheric CO_2 measurements are very important for understanding the carbon cycle because CO_2 mixing ratios in the atmosphere are strongly affected by photosynthesis, respiration, oxidation of organic matter, biomass and fossil fuel burning, and air–sea exchange process (Machida et al., 2003).

The present study brings out first continuous measurements of atmospheric GHG's using high precision Los Gatos Research's–greenhouse gas analyser (LGR-GGA) over Shadnagar, a suburban site of Central India during the period 2014. In addition to GHG's observations, we have also made use of an automatic weather station (AWS) data along with model/satellite retrieved observation during the study period. Details about study area and data sets are described in the following sections.

2 Study area

Shadnagar is situated in Mahabubnagar district of newly formed Indian state of Telangana. It is a rural location situated ~ 70 km away from urban site of Hyderabad (Northern side) with a population of ~ 0.158 million (Patil et al., 2013). A schematic map of study area is shown in Fig. 1a. Major source of pollutants over Shadnagar can be from small and medium scale industries, biomass burning and bio-fuel as well as from domestic cooking. In the present study sampling of GHG's and related meteorological parameters are carried out in the premises of National Remote Sensing Center (NRSC), Shadnagar Campus (17°02' N, 78°11' E). Sampling site is near to National highway 7 (NH7) and a railway track (non-electrified) is in the East (E) direction.

Mean monthly variations of temperature (°C) and RH (%) observed at Shadnagar during 2014 are shown in Fig. 1b and c respectively. The Indian Meteorological Department (IMD) defined monsoon as June–July–August–September (JJAS), post-monsoon (October–November–December – OND), winter (January–February – JF) and pre-monsoon (March–April–May – MAM) in India. Temperature over Shadnagar varies from ~ 20 to ~ 29°C. Relative humidity (RH) in Shadnagar reached a maximum of 82 % in monsoon from a minimum of 48 % recorded during pre-monsoon. Surface wind speed (Fig. 1d) varies between 1.3 to 1.6 ms⁻¹ with a maximum observed during monsoon and minimum in pre-monsoon. The air mass advecting (Fig. 1e) towards study site is either easterly or westerly. The easterly wind prevails during winter and gradually shifts to south-westerlies in pre-monsoon, and dominates during monsoon.

3 Data set and methodology

Details about the instrument and data utilized are discussed in this section. The availability and frequency of the observations all data used in present study are tabulated in Table 1.

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operating principle and has an in-built calibration unit for conducting periodical span and zero checks. The NO_x analyzer utilizes a molybdenum converter to convert NO₂ into NO and estimates the NO_x concentration by the intensity of light emitted during the chemiluminescent reaction of NO with O₃ present in the ambient air. The analyzer is integrated with zero and span calibration which are performed twice monthly.

Simultaneous observations of meteorological parameters are obtained from an automatic weather stations (AWS) located in the same campus.

3.2 Satellite and model observations

3.2.1 MODIS

Moderate-resolution Imaging Spectrometer (MODIS) is launched in December 1999 on the polar-orbiting NASA-EOS Terra platform (Salomonson et al., 1989; King et al., 1992). It has 36 spectral channels and acquires data in 3 spatial resolutions of 250, 500 m, and 1 km (channels 8–36), covering the visible, near-infrared, short-wave infrared, and thermal-infrared bands. In the present study we used monthly Normalised Difference Vegetation Index (NDVI) data obtained from Terra/MODIS at 5 km spatial resolution. The NDVI value is defined as following ratio of albedos (α) at different wavelengths:

$$\text{NDVI} = \frac{\alpha_{0.86\mu\text{m}} + \alpha_{0.67\mu\text{m}}}{\alpha_{0.86\mu\text{m}} - \alpha_{0.67\mu\text{m}}} \quad (1)$$

NDVI values can range from –1.0 to 1.0 but typical ranges are from 0.1 to 0.7, with higher values associated with greater density and greenness of plant canopies. More details of the processing methods used in generating the data set can be found in James and Kalluri (1994).

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3.2.2 COSMIS-RO

COSMIC (Constellation Observation System for Meteorology, Ionosphere and Climate) is a GPS (Global Positioning System) radio occultation (RO) observation system (Wang et al., 2013). It consists of six identical microsattellites, and was launched successfully on 14 April 2006. GPS radio occultation observation has the advantage of near-global coverage, all-weather capability, high vertical resolution, high accuracy and self-calibration (Yunck et al., 2000). Geophysical parameters like temperature and humidity profiles have been simultaneously obtained from refractivity data using one-dimensional variational (1DVAR) analysis. Further COSMIC-RO profiles are used to estimate planetary boundary layer height (BLH). BLH is defined to be the height at which the vertical gradient of the refractivity or water vapor partial pressure is minimum (Ao et al., 2012), explained detail methodology for calculating the BLH from refractivity (N).

3.2.3 Hysplit model

The general air mass pathway reaching over Shadnagar is analysed using HYSPLIT model (Draxler and Rolph, 2003) (<http://www.arl.noaa.gov/ready/hysplit4.html>). We computed 5 day isentropic model backward air mass trajectory for all study days with each trajectory starting at 06:00 UTC and reaching study site, (Shadnagar) at different altitudes(1, 2, 3 and 4 km). Even though the trajectory analysis have inherent uncertainties (Stohl, 1998), they are quite useful in determining long range circulation.

4 Results and discussion

4.1 Seasonal variations of CO₂ and CH₄

Monthly variations of CO₂ and CH₄ during the study period are shown in Fig. 2a and b. Annual mean of CO₂ over study region is found to be $394 \pm 2.92 (\mu \pm 1\sigma)$ ppm with

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an observed minimum in monsoon and maximum in pre-monsoon. Background (average) values of CO_2 observed during different seasons are 393 ± 5.60 , 398 ± 7.60 , and 392 ± 7.0 and 393 ± 7.0 ppm with respectively winter, pre-monsoon, monsoon and post-monsoon. Minimum CO_2 during winter (dry season) indicates the loss of carbon (Gilmanov et al., 2004) as decreased temperature and solar radiation during this period inhibit increases in local CO_2 assimilation (Thum et al., 2009). Enhancement in Pre-monsoon is due to higher temperature and solar radiation prevailing during these months which stimulate the assimilation of CO_2 in the daytime and respiration in the night (Fang et al., 2014). Surface CO_2 concentration recorded a minimum during monsoon months can be mainly because of enhanced photosynthesis processes with the availability of greater soil moisture (Patil et al., 2013). Further increase during post-monsoon CO_2 is associated with high ecosystem productivity (Sharma et al., 2014) also an enhancement in soil microbial activity (Kirschke et al., 2013).

CH_4 concentration in the troposphere is principally determined by a balance between surface emission and destruction by hydroxyl radicals (OH). The major sources for CH_4 in the Indian region are rice, paddies, wetlands and ruminants (Schneising et al., 2009). Annual CH_4 concentration over study area is observed to be 1.92 ± 0.07 ppm, with a maximum (2.02 ± 0.01 ppm) observed in post-monsoon and minimum (1.85 ± 0.03 ppm) in monsoon. The highest concentration appears during post-monsoon and may be associated with the Kharif season (Goroshi et al., 2011). Background (average) values of CH_4 observed during different seasons are 1.93 ± 0.05 , 1.89 ± 0.05 , 1.85 ± 0.03 and 2.02 ± 7 ppm with respectively winter, pre-monsoon, monsoon and post-monsoon. Low mixing ratios of CH_4 observed during monsoon season were mainly due to the reduction in atmospheric hydrocarbons because of the reduced photochemical reactions and the substantial reduction in solar intensity (Gaur et al., 2014). The rate of change of CH_4 was found to be high during post-monsoon and winter. Both biological and physical processes control the exchange of CH_4 between rice paddy fields and the atmosphere. This may be one of the major reasons for the

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perature does not significantly influence seasonal variation of CH_4 (Chen et al., 2015). Seasonal variation of GHG's also showed an insignificantly negative correlation with relative humidity. A similar observation is also reported by Abhishek et al. (2014). One of the supporting argument can be in humid conditions, these stoma can fully open to increase the uptake of CO_2 without a net water loss. Also, wetter soils can promote decomposition of dead plant materials, releasing natural fertilizers that help plants grow.

Influence of boundary layer height on GHGs mixing ratios

The planetary boundary layer is the lowest layer of the troposphere where wind speed as a function of temperature plays major role in its thickness variation. It is an important parameter for controlling the observed diurnal variations and potentially masking the emissions signal (Newman et al., 2013). Since complete set of COSMIC RO data is not available during the study period, in this analysis we have analysed RO data from July 2013 to June 2014, along with simultaneous observations of GHG's. Monthly variations (Figure not show) of BLH computed from high vertical resolution of COSMIC-RO data against CO_2 and CH_4 concentrations. Monthly BLH is observed to be minimum (maximum) during winter and monsoon (pre monsoon) seasons. The highest (lowest) BLH over study region was identified 3.20 km (1.50 km). An average monthly air temperature is maximum (minimum) of 29°C (20°C) during summer (winter) months.

Seasonal change in BLH thickness over study region was observed to be as Monsoon (M, 1.74 km) < winter (W, 2.10 km) < Post Monsoon (PM, 2.30 km) < Pre-monsoon (Pre-M, 3.15 km); its influence on CO_2 and CH_4 mixing ratios are shown in Fig. 5a and b. As seasonal BLH thickness increase, mixing ratios of CO_2 (CH_4) decreased from 8.68 to 5.86 ppm (110 to 40 ppb). The amount of biosphere emissions influence on CO_2 and CH_4 can be estimated through atmospheric boundary layer processes. Since the study region being a flat terrain variations in CO_2 and CH_4 were mostly influenced by boundary layer thickness through convection and biosphere activities.

4.4 Correlation between CO₂ and CH₄

A correlation study is carried out between hourly averaged CO₂ and CH₄ during all season for the entire study period. The statistical analysis for different seasons is shown in Table 3. Fang et al. (2015) suggest that correlation coefficient (R) value higher than 0.50 indicates similar source mechanism of CO₂ and CH₄. Also, a positive correlation dominance of anthropogenic emission on carbon cycle. Our study also reveals a strong positive correlation observed between CO₂ and CH₄ during winter, pre-monsoon, monsoon and post-monsoon with R equal to 0.80, 0.80, 0.61 and 0.72 respectively. Seasonal regression coefficients (slope) and their uncertainties (ψ_{slope} , $\psi_{y\text{-int}}$) are computed using Taylor (1997) which showed maximum during winter, pre-monsoon and minimum in monsoon that figure out the hourly stability of the mixing ratios between CO₂ and CH₄. This can be due to relatively simple source/sink process of CO₂ in comparison with CH₄. Dilution effects during transport of CH₄ and CO₂ can be minimized to some extent by dividing the increase of CH₄ over time by the respective increase in CO₂ (Worthy et al., 2009). Figure 6 shows the seasonal variation of $\Delta\text{CH}_4/\Delta\text{CO}_2$. In this study, background concentration of respective GHG's are determined as mean values of the 1.25 percentile of data for monsoon, post-monsoon, pre-monsoon and winter (Pan et al., 2011; Worthy et al., 2009). Annual $\Delta\text{CH}_4/\Delta\text{CO}_2$ over the study region during the study period is found to be 7.1 (ppb ppm⁻¹). This low value clearly indicates the dominance of CO₂ over the study region. The reported $\Delta\text{CH}_4/\Delta\text{CO}_2$ values from some of the rural sites viz Canadian Arctic and Hateruma Island (China) is of the order 12.2 and ~ 10 ppb ppm⁻¹ respectively (Worthy et al., 2009; Tohjima et al., 2014). Average $\Delta\text{CH}_4/\Delta\text{CO}_2$ ratio during winter, pre-monsoon, monsoon and post-monsoon are 9.40, 6.40, 4.40, and 8.20 ppb respectively. Monthly average, of $\Delta\text{CH}_4/\Delta\text{CO}_2$, is relatively high from late post-monsoon to winter, when the biotic activity is relatively dormant (Tohjima et al., 2014). During pre-monsoon decrease in $\Delta\text{CH}_4/\Delta\text{CO}_2$ ratio indicates the enhancement of CO₂ relative to that of CH₄.

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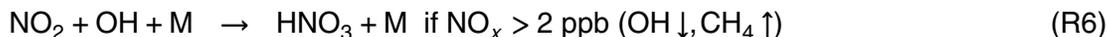


4.5 Methane (CH₄) sink mechanism

Methane (CH₄) is the most powerful greenhouse gas after CO₂ in the atmosphere due to its strong positive radiative forcing (IPCC, 1990; Stocker et al., 2013). Atmospheric CH₄ is mainly (70–80 %) from biological origin produced in anoxic environments, by anaerobic digestion of organic matter (Crutzen and Zimmermann, 1991). The major CH₄ sink is oxidation by hydroxyl radicals (OH), which accounts for 90 % of CH₄ sink (Vaghjiani and Ravishankara, 1991; Kim et al., 2015). OH radicals are very reactive and are responsible for the oxidation of almost all gases in the atmosphere. Primary source for OH radical formation in the atmosphere is photolysis of ozone (O₃) and water vapor (H₂O). Eisele et al. (1997) defined primary and secondary source of OH radicals in the atmosphere. Primary source of OH radical is as follows;



Removal of CH₄ is constrained by the presence of OH radicals in the atmosphere. A 1 min time series analysis of CH₄, NO_x, O₃ and H₂O and associated wind vector for August 2014 to understand the CH₄ chemistry is shown in Fig. 7a and b. Low NO_x (1–2 ppb) values are shown in horizontal elliptical region of Fig. 7a and observed corresponding low CH₄ (1.80 ppm) concentrations. The low NO_x in turn produces high OH radicals in the atmosphere due to conversion of HO₂ radical by NO, which removes CH₄ through oxidation process as shown below.



Crutzen and Zimmermann (1991) and Eisele et al. (1997) observed that at low NO_x (0.5–2.0 ppb) levels most HO_x family radicals such as HO₂ and peroxy radicals (RO₂)

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Table 2. Seasonal amplitudes of CO₂ and CH₄ over study region arriving from different directions.

Wind Direction	Winter $\frac{\text{CO}_2}{\text{CH}_4}$ (ppm)	Pre-monsoon $\frac{\text{CO}_2}{\text{CH}_4}$ (ppm)	Monsoon $\frac{\text{CO}_2}{\text{CH}_4}$ (ppm)	Post-monsoon $\frac{\text{CO}_2}{\text{CH}_4}$ (ppm)
0–45	399.85/1.98	410.37/1.94	400.72/1.91	395.13/2.02
45–90	391.66/1.94	399.59/1.89	388.82/1.91	390.23/1.98
90–135	391.57/1.93	397.79/1.87	388.99/1.87	389.06/1.97
135–180	389.34/1.89	393.87/1.85	391.81/1.86	387.69/1.97
180–225	391.14/1.89	396.75/1.85	390.28/1.82	392.30/2.02
225–270	389.13/1.88	394.81/1.86	390.26/1.82	384.40/1.94
270–315	388.68/1.87	398.68/1.89	389.58/1.82	384.99/1.93
315–360	390.87/1.91	401.17/1.89	387.58/1.83	389.32/1.98

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Table 3. Statistical correlation between CO₂ and CH₄.

S.No	Seasons	Correlation coefficient (R^2)	Slope ($\frac{Y_{CH_4} \text{ (ppm)}}{X_{CO_2} \text{ (ppm)}}$)	ψ_{slope} (ppm)	$\Psi_{y\text{-int}}$ (ppm)
1	Monsoon (JJAS)	0.37	0.005	0.00015	1.91
2	Post-monsoon (OND)	0.52	0.0065	0.00014	1.52
3	Winter (JF)	0.61	0.0085	0.00018	9.13
4	Pre-monsoon (MAM)	0.64	0.0059	0.00021	2.73

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Table 4. Cluster analysis of air mass trajectories reaching Shadnagar at various heights during different seasons.

Seasonal Backward trajectory (%)	NW				NE				SE				SW			
	1 km	2 km	3 km	4 km	1 km	2 km	3 km	4 km	1 km	2 km	3 km	4 km	1 km	2 km	3 km	4 km
Winter	54	32	2	0	32	24	44	52	10	25	11	7	4	19	42	41
Pre-monsoon	24	9	8	1	26	31	64	78	36	46	2	10	14	14	26	11
Monsoon	0	1	7	19	12	34	80	70	4	4	4	6	84	61	9	5
Post-monsoon	42	15	11	14	47	53	41	49	8	30	32	26	3	2	16	11

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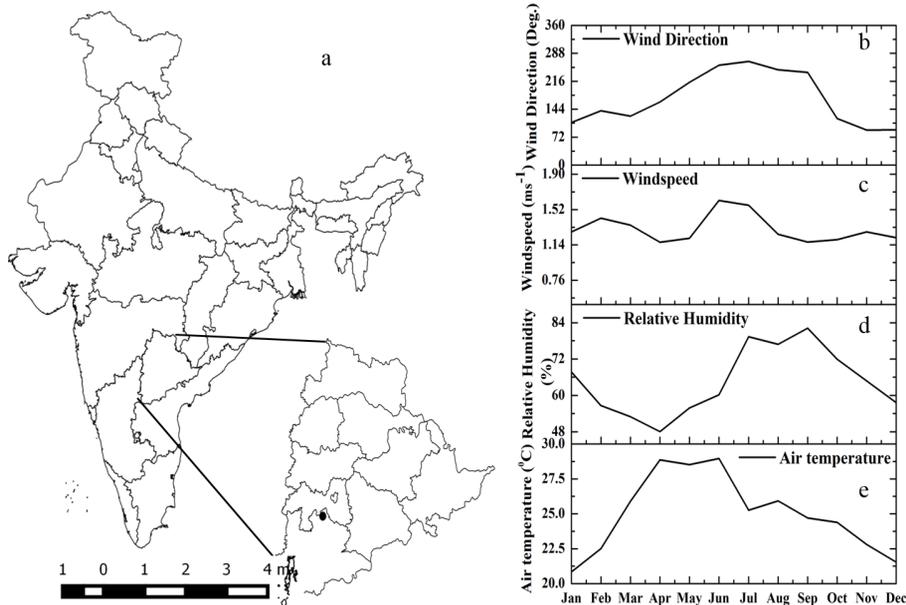


Figure 1. (a) Schematic representation of study area; (b–e) Seasonal variation of prevailing meteorological conditions during study period.

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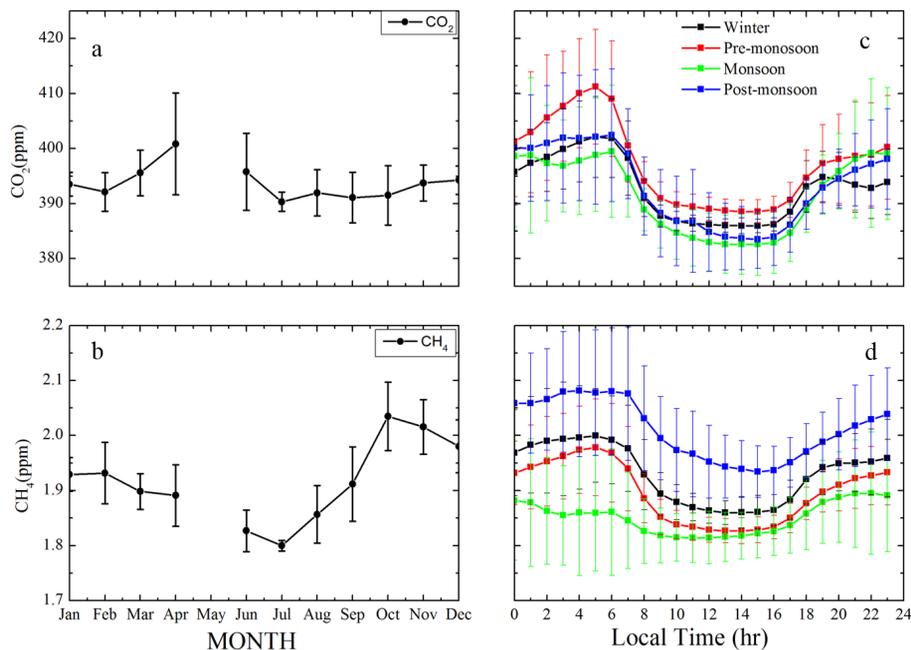


Figure 2. (a–b) Seasonal variations of CO₂ and CH₄; (c–d) diurnal variations of CO₂ and CH₄ during 2014.

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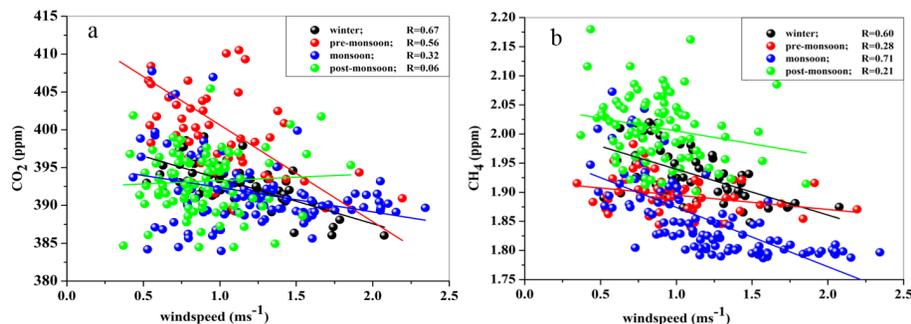


Figure 3. (a–b) Scatterplot between wind speed and GHGs (CO₂ and CH₄).

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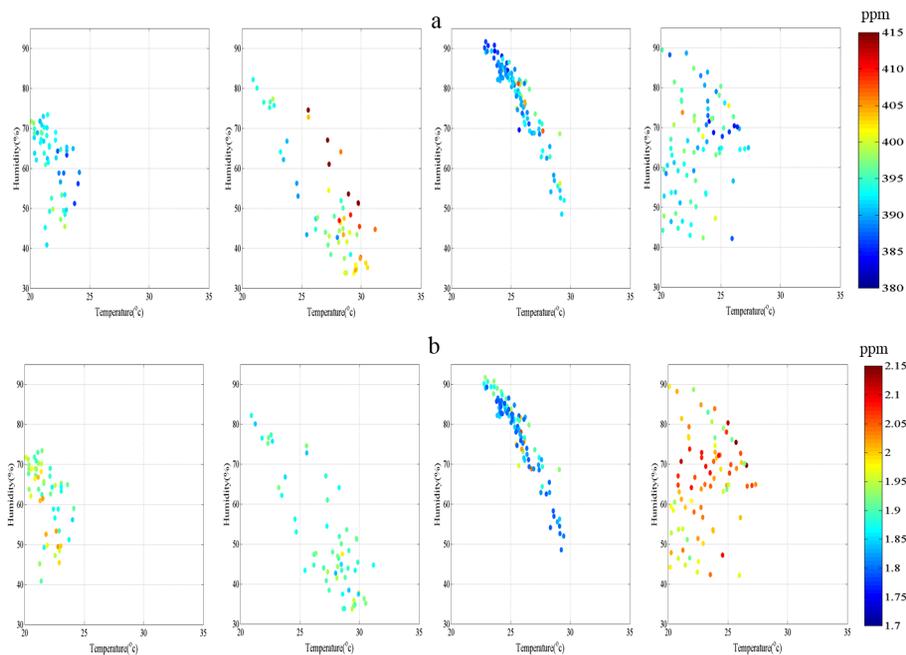


Figure 4. (a) Seasonal variation of CO₂ as function of humidity and temperature during winter, pre-monsoon, monsoon and post-monsoon. (b) Seasonal variation of CH₄ as function of humidity and temperature during respective seasons.

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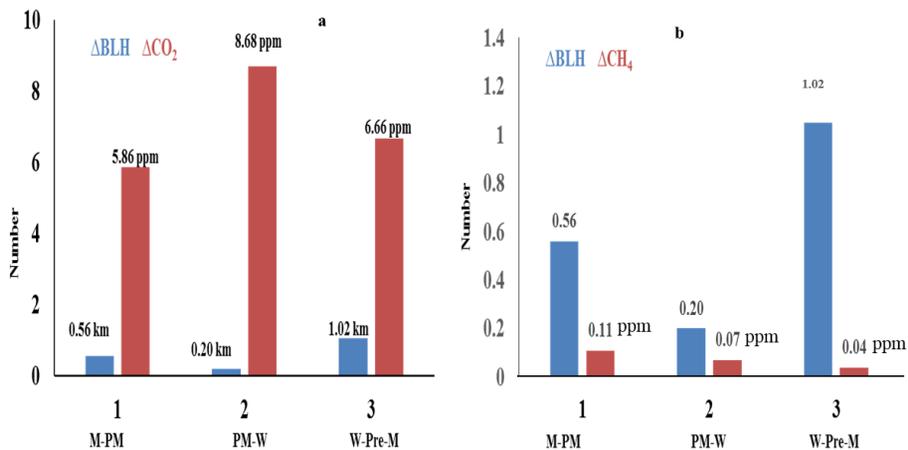


Figure 5. Seasonal variations of (a) CO_2 and (b) CH_4 against boundary layer height change.

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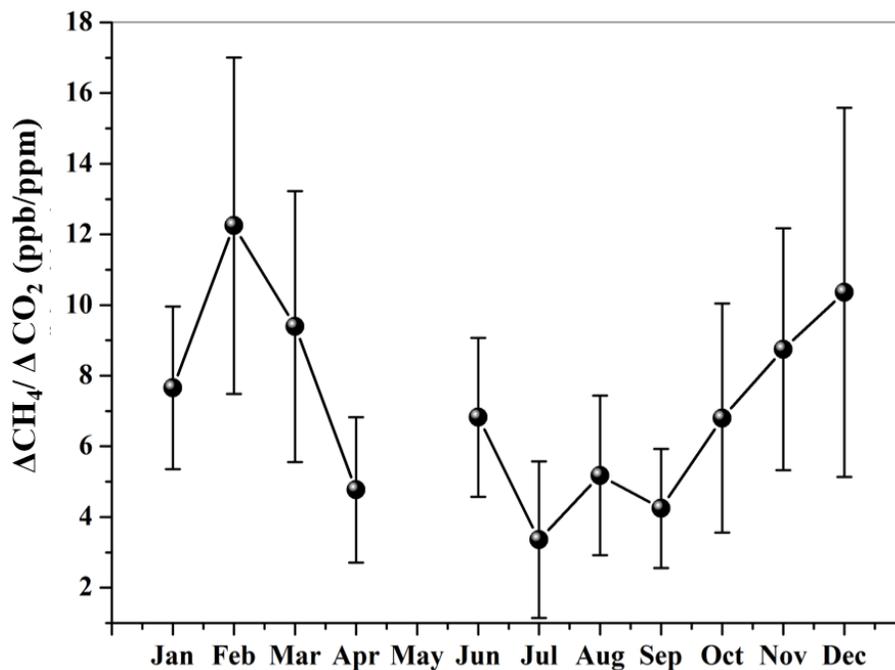


Figure 6. Monthly variation of $\Delta\text{CH}_4/\Delta\text{CO}_2$ during study period.

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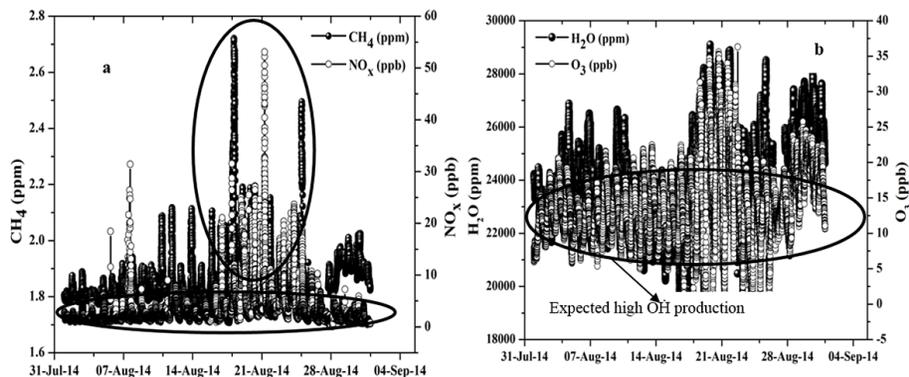


Figure 7. Time series analysis of (a) CH₄ vs. NO_x, (b) H₂O vs. O₃.

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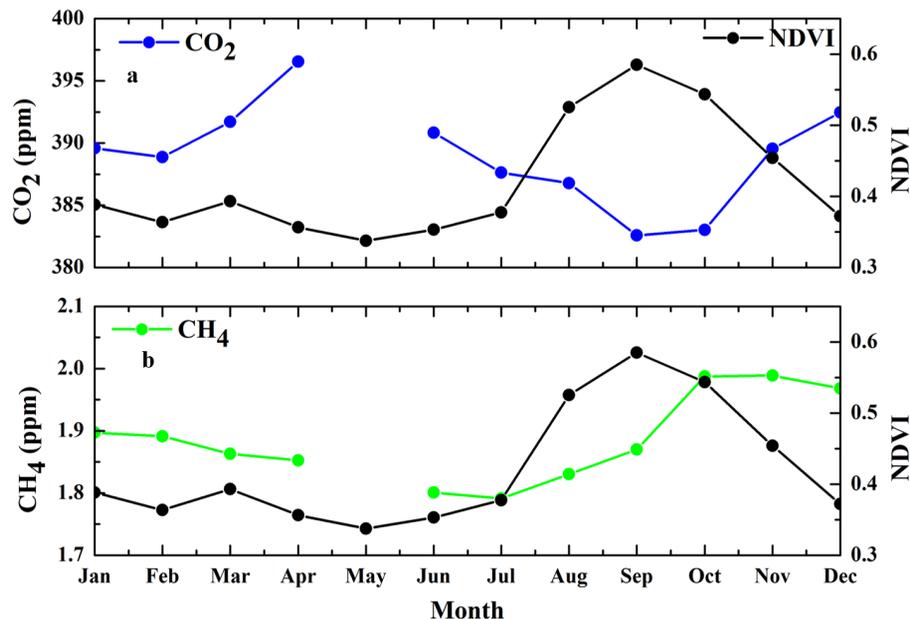


Figure 8. (a) Seasonal variation of CO₂ in conjunction with NDVI (Normalized Difference Vegetation Index). (b) Seasonal variation of CH₄ in conjunction with NDVI.

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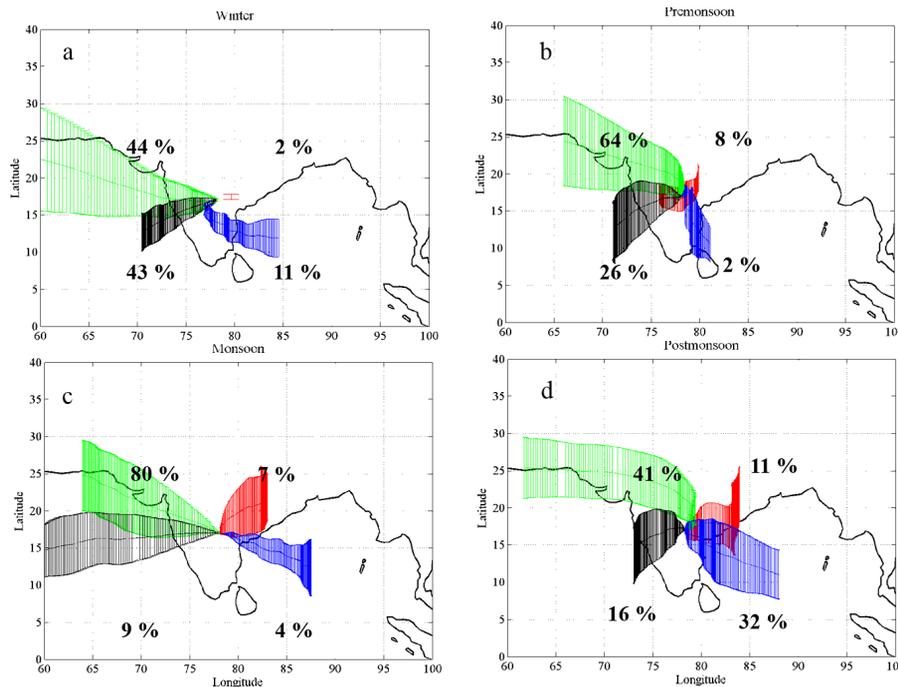


Figure 9. (a–d) Long range circulation of air mass trajectories ending over Shadnagar at 3 km during winter, pre-monsoon, monsoon and post-monsoon.

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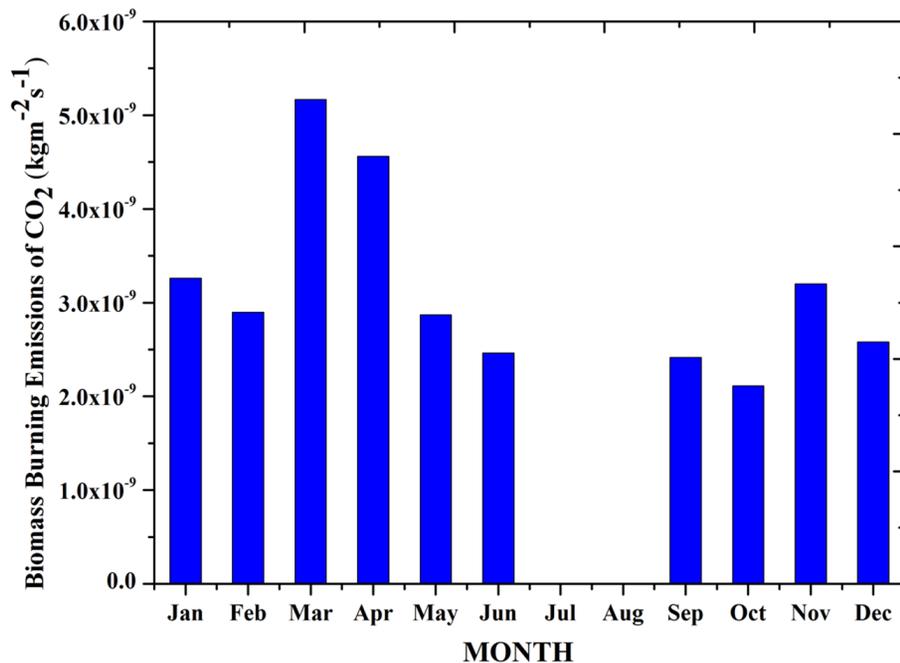


Figure 10. Long term analysis of CO₂ biomass burning emissions over study region.

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