Dear Referees,

Thank you very much for your valuable suggestions and comments. We have tried to implement each point by point. Please see the blue colored text is answers to the query. Detailed explanations with updated figures have been incorporated in the revised manuscript.

Referee#1:

General comments: Atmospheric greenhouse gases (GHGs) are important climate forcing agents and have significant impacts on global climate. This study brings outs first continuous measurements of atmospheric GHGs (CO_2 and CH_4) using high precision Los Gatos Research's-greenhouse gas analyser (LGR-GGA) over Shadnagar, a suburban site of Central India during the period of 2014. The authors also investigate the influences of meteorology on GHGs and their interrelationship. It is useful to estimate quantitatively the radiative effects of GHGs on regional or global climate change. Obviously, there are numerous grammatical and technical errors in the manuscript. This paper is reconsidered to be acceptable and published after major revisions.

Specific comments:

Q1). Abstract, lines 1-18: "Atmospheric greenhouse gases (GHGs) such as carbon dioxide: : :: : :It implies the seasonal variations in source-sink mechanisms of CO_2 and CH_4. Present study also confirms implicitly the presence OH radicals as a major sink of CH_4 over the study region". âG˘ Š This study aims to analyse the seasonal variations of CO_2 and CH_4 over a suburban site of Central India and investigate the influences of prevailing meteorology (e.g., air temperature, wind speed, wind direction, relative humidity, boundary layer height) on GHGs and their interrelationship. The manuscript also reveals that biomass burning (forest fire and crop residue burning) has a role in pre-monsoon enhancement of CO_2 over study site (Page 34219, lines 4-17). And the air mass trajectories of crops agriculture residue burning in the NW and NE regions part of India can reach the study site at different altitudes during post-monsoon to early pre-monsoon (Page 34220, lines 1-5). Therefore, in order to investigate the exact influences of prevailing meteorology on GHGs, the CO_2 contributions of regional biomass burning and long range transport should be excluded from the CO_2 measurements. Otherwise, many conclusions in present study make no sense.

Answer: We agree with referee suggestions. Inorder to investigate the exact influences of prevailing meteorology on GHGs, we have done the analysis, by eleminating those study days where biomass burnig influence are there. Biomass burning days are identified using MODIS active fire count data during the study period. Figure 3a-c is updated accordingly.

Q2). "4.6 Influence of vegetation on GHGs", Page34218, lines 25-28 and Page34219, lines 1-3: "The main source for CH_4 emissions are soil microbial (Kirschke et al., 2013) activity which are more active during monsoon and post monsoon seasons: : :: : :The predominating factors which control the soil emissions of CO_2 and CH_4 are moisture content, soil temperature, vegetation and soil respiration (Smith et al., 2003; Jones et al., 2005; Chen et al., 2010) respectively." âG˘ Š So, we recommend strongly that the authors add and discuss the possible influences of soil parameters (such as, moisture content, soil temperature, vegetation and soil respiration) on GHGs and their interrelationship in the manuscript. And we believe that the authors would acquire many interesting findings.

Answer: As per referee suggestion we have studied the influence of soil moisture and temperature on GHGs. A new figure (Figure 8) is added illustrating the influence of of soil moisture and temperature on GHGs and corresponding discussion are provided in the manuscript.

Q3). Page 34212, lines 6-9: "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)" âG˘ Š As discussing in the later section (4.6 and 4.7), quantitative contributions of regional biomass burning and long range transport to atmospheric CO_2 concentration at the study site are very important to the interpretation of enhancement CO_2 in pre-monsoon.

Answer: A quantative analysis (in terms of a case study) of regional/long range transported influence of biomass burning on local GHGs concencetration is provided in section 4.2.

Q4). Figure 3 to Figure 5: The authors study the interrelationships between monthly mean meteorology and GHGs. We suggest that the authors add the interrelationships between daily mean meteorology and GHGs, whether it is the same variation with monthly mean? If not, what is about for daily average?

Answer: Figure 3 and 4 updated as figure 6 to figure 8 and figure 5 as figure 9 in the revised manuscript. In figure 6-8, interrelationships are analysed using daily mean data. Each points in the Figure 6-8 represent daily mean corresponding to each season. Howerver for Figure 9 (Seasonal variation of GHGs and Boundary layer height) daily data on boundary layer height over study region is not available from satellite, we used ECMWF-ERA data to study the diurnal boundary layer (figure 5) effect on GHGs mixing ratios over study region.

Minor Comments:

Comment: 1

Title: "Influence of meteorology and interrelationship with greenhouse gases (CO_2 and CH_4) at a sub-urban site of India" âG˘ Š Change "sub-urban" to "suburban", and modify the other places in the whole manuscript.

Answer: The correction is incorporated in the revised manuscript.

Comment: 2

Abstract, Page 34206, lines 2-4: "Atmospheric greenhouse (GHGs) such as carbon dioxide (CO_2) and methane (CH_4) are important climate forcing agents due to their significant impact on the climate system." âG˘ Š Change to "Atmospheric greenhouse (GHGs), such as carbon dioxide (CO_2) and methane (CH_4), are important climate forcing agents due to their significant impacts on the climate system."

Answer: The correction is incorporated in the revised manuscript

Comment: 3

Abstract, Page 34206, line 5; Page 34207, line 24; Page 34208, line 7: change “GHG’s” to “GHGs”, and modify the other places in the whole manuscript.

Answer: updated in the revised manuscript

Comment: 4

Abstract, Page 34206, lines 6-8; Page 34220, lines 10-11: “The annual mean of CO2 and CH4 over the study region is found to be 394_2.92 and 1.92_0.07 ppm (mean, ïA¸ _1SD, ïA¸ s¸) respectively.” âG˘ Š Change to “The annual mean CO2 and CH4 over the study region are found to be 394_2.92 ppm and 1.92_0.07 ppm (mean_standard deviation, ïA¸ _1ïA¸ s¸) respectively.”

Answer: The correction is incorporated in the revised manuscript.

Comment: 5

Abstract, Page 34206, line 8, line 14; Page 34220, line 11: change “showed” to “show” and keep the consistency in the manuscript.

Answer: Corrected as suggested.

Comment: 6

Abstract, Page 34206, lines 14-16: “CO2 and CH4 showed a strong positive correlation during winter, pre-monsoon, monsoon and post-monsoon with R equal to 0.80, 0.80, 0.61 and 0.72 respectively.” âG˘ Š Change to “CO2 and CH4 show a strong positive correlation during winter, pre-monsoon, monsoon, and post-monsoon with correlation coefficients (Rs) equal to 0.80, 0.80, 0.61, and 0.72, respectively.”

Answer: The correction is incorporated in the revised manuscript.

Comment: 7

“Abstract, Page 34206, lines 17-18: “Present study also confirms implicitly the presence OH radicals as a major sink of CH4 over the study region.” âG˘ Š Change to “Present study also confirms implicitly the presence hydroxyl radicals (OH) as a major sink of CH4 over the study region.”

Answer: Updated in the revised manuscript.

Comment: 8

Page 34206, line 21: change “globalwaming” to “global warming”

Answer: Updated in the revised manuscript.

Comment: 9

Page 34206, lines 22-24: “CO_2 and CH_4 concentrations have increased by 40 and 150âG˘ Š Change to “CO_2 and CH_4 concentrations have increased by 40 Please add the citation of Huang et al., 2015 in the manuscript: Huang J.*, Yu H., Guan X., Wang G. and Guo R., 2015: Accelerated dryland expansion under climate change, Nature Climate Change, doi:10.1038/nclimate2837.

Answer: Thanks to reviewer for providing very useful information and suggestions about the GHGs variability of arid and semiarid areas. It helped to improve the manuscript. We have included Huang et. al (2015) reference in the manuscript.

Reference: Huang, J., Yu, H., Guan, X., Wang, G., & Guo, R. (2015). Accelerated dryland expansion under climate change. Nature Climate Change, doi:10.1038/nclimate2837.

Comment: 10

Page 34207, line 3: change “constitutes” to “constitute”

Answer: Updated in the revised manuscript.

Comment: 11, 12, 13, and 14

Page 34207, line 6: delete “that” (12) Page 34207, line 8: change “andecosystems” to “and ecosystems” (13) Page 34207, line 13: change “the part of the atmosphere” to “part of the atmosphere” (14) Page 34207, line 14: change “donimatethe” to “dominate the”

Answer: Updated in the revised manuscript.

Comment: 15

Page 34208, lines 5-7: “Major source of pollutants over Shadnagar can be from small and medium scale industries, biomass burning and bio-fuel aswell as from domestic cooking.” âG˘ Š Change to “Major sources of pollutants over Shadnagar can be from small and medium scale industries, biomass burning and bio-fuel as well as from domestic cooking.”

Answer: Incorporated in the revised manuscript.

Comment: 16

Page 34208, lines 11-12: “Mean monthly variations of temperature (â˘D ˘ C) and RH (â ˘ GŠ Change to “Monthly mean variations of temperature (â˘DC˘) and relative humidity (RH,

Answer: Updated in the manuscript.

Comment: 17

Page 34208, line 16: Change “Relative humidity (RH) in Shadnagar reached a maximum of 82

Answer: Updated in the manuscript.

Comment: 18

Page 34209, lines 17-21: “In the present study we used GGA retrieved CO2 and CH4 data. High resolution data are diurnally averaged and is used in further analysis. Due to failure of internal central processing unit (CPU) of the analyzer data is not recorded from pre-monsoon month of May to a few days in June during the study period.” âG˘ Š Change to “In the present study we used GGA to retrieve CO2 and CH4 data. High resolution data sets are diurnally averaged and used in further analysis. Due to failure of internal central processing unit (CPU) of the analyzer, data are not recorded from pre-monsoon month of May to a few days in June during the study period.”

Answer: The correction is incorporated in the revised manuscript.

Comment: 19

Page 34209, lines 23-25: “Surface concentrations of O3 and NOX have been measured continuously using on-line analyzers Model No.s: 49i and 42i for O3 and NOX respectively, procured from Thermo Scientific, USA) since July 2014.” âG˘ Š Change to “Surface concentrations of O3 and NOX have been measured continuously using on-line analyzers (Model No.s: 49i and 42i for O3 and NOX respectively), procured from Thermo Scientific, USA since July 2014.”

Answer: The brackets modified in the revised manuscript.

Comment: 20

Page 34210, lines 2-3: “The NOx analyzer utilizes a molybdenumconverter to convert NO2 into NO and estimates the NOx concentration: : :” âG˘ Š Change to “The NOx analyzer utilizes a molybdenum converter to convert NO2 into NO and estimate the NOx concentration: : :”

Answer: Incorporated in the revised manuscript.

Comment: 21, 22, & 23

Page 34210, line 5: change “zero and span calibration” to “zero and span calibrations” (22) Page 34210, line 7: change “an automatic weather stations (AWS)” to “an automatic weather station (AWS)” (23) Page 34210, line 13: change “of 250, 500 m, and 1 km” to “of 250 m, 500 m, and 1 km”

Answer: Incorporated in the revised manuscript.

Comment: 24

Page 34211, lines 7-8: ”Geophysical parameters like temperature and humidity profiles have been simultaneously obtained from: : :” âG˘ Š Change to ”Geophysical parameters (such as, temperature and humidity profiles) have been simultaneously obtained from: : :”

Answer: Updated in the revised manuscript.

Comment: 25 & 26

Page 34212, lines 1-2: “Background (average) values of CO_2: : :” âGˇ Š Please add “How to define or calculate background average values of CO_2: : :” (26) Page 34212, line 3: change “and 392_7.0 and 393_7.0 ppm with respectively winter, pre-monsoon, monsoon and post-monsoon.” to “392_7.0, and 393_7.0 ppm respectively with winter, pre-monsoon, monsoon, and post-monsoon.”

Answer: The correction is incorporated in the revised manuscript.

Comment: 27, 28, and 29

Page 34213, line 4: change “Figure 2c and d depicts” to “Figure 2c and 2d depict” (28) Page 34213, line 6: change “such as land use land cover change” to “such as land use and land cover change” (29) Page 34213, line 26: change “while a not so significant correlation suggest the influence of regional transport” to “while a not so significant correlation suggests the influence of regional transport”

Answer: Updated in the manuscript.

Comment: 30

Page 34214, lines 1-5: “Figure 3a and b shows scatter plot between GHG’s and wind speed during different seasons. Analysis of Fig. 3b shows that there exist an inverse correlation between monthly mean wind speed and GHG’s. Correlation coefficient (R) between wind speed and CO_2 during pre-monsoon, monsoon, post-monsoon and winter is 0.56, 0.32, 0.06 and 0.67 respectively.” âGˇ Š Change to “Figure 3a and 3b show scatter plot between GHG’s and wind speed during different seasons. Fig. 3b shows that there exists an inverse correlation between monthly mean wind speed and GHG. Correlation coefficients (Rs) between wind speed and CO_2 during pre-monsoon, monsoon, post-monsoon, and winter are 0.56, 0.32, 0.06, and 0.67, respectively.”

Answer: Incorporated in the revised manuscript.

Comment: 31, 32, 33, 35 and 36

Page 34214, line 16: change “The meteorological parameters (temperature and relative humidity) influenceon trace gases” to “The influence of meteorological parameters (temperature and relative humidity) on trace gases” (33) Page 34214, line 18: change “shows the scatter plot of temperature vs. relative humidity” to “show the scatter plot of temperature versus relative humidity” (34) Page 34214, line 19: change “Hence, dailymean data” to “Hence, daily mean data” (35) Page 34215, line 17: change “An average monthly air temperature” to “A monthly average air temperature”

Answer: Incorporated in the revised manuscript.
Comment: 36

Page 34216, line 1: change “between hourly averaged CO_2 and CH_4 during all season” to “between hourly average CO_2 and CH_4 during all seasons”

Answer: Updated in the revised manuscript.

Comment: 37

Page 34216, line 21: change "is" to "are"

Answer: Changed as suggested.

Comment: 38

Page 34217, lines 3-6: "Atmospheric CH_4 is mainly (70-80âG˘ Š Hence, it is very essential to discuss the possible influences of soil parameters (such as, moisture content, soil temperature, vegetation and soil respiration) on GHGs and their interrelationship in the manuscript.

Answer: We updated in the revised manuscript at section 4.5.

Comment: 39

Page 34218, lines 20-21: change "..is calculated from daily day time (10:00-16:00 LT) mean." to "..is calculated from daily mean in day time (10:00-16:00 LT)."

Answer: Incorporated in the revised manuscript.

Comment: 40

Page 34219, line 19: change "To understand the role of long range circulation we separated the trajectory into 4 clusters" to "To understand the role of long range circulation, we separated the trajectory into 4 clusters"

Answer: Updated in the manuscript.

Comment: 41

Page 34220, lines 20-22: "Correlation coefficient (R) between wind speed and CO2 during pre-monsoon, monsoon, post-monsoon and winter is 0.56, 0.32, 0.06 and 0.67 respectively. While for CH4 it is found to be 0.28, 0.71, 0.21, and 0.60 respectively." âG˘ Š Change to "Correlation coefficients (Rs) between wind speed and CO2 during pre-monsoon, monsoon, post-monsoon and winter are 0.56, 0.32, 0.06, and 0.67, respectively. While CH4 are found to be 0.28, 0.71, 0.21, and 0.60, respectively."

Answer: Incorporated in the revised manuscript.

Comment: 42

Page 34232: "Figure 1. b to e represent the seasonal variations of wind direction, wind speed, relative humidity, and air temperature.", please keep the consistencies with the context (Page 34208, line 11; lines 18-19). And please add the different symbols of monthly mean variations of prevailing meteorology from Fig. 1b to 1e.

Answer: Figure 1 is updated as referee suggested.

Comment: 43

The following related citations are recommended to be quoted in the manuscript: [1] Huang, J.*, W. Zhang, J. Zuo, J. Bi, J. Shi, X. Wang, Z. Chang, Z. Huang, S. Yang, B. Zhang, G. Wang, G. Feng, J. Yuan, L. Zhang, H. Zuo, S. Wang, C. Fu and J. Chou, 2008: An overview of the Semi-Arid Climate and Environment Research Observatory over the Loess Plateau, Advances in Atmospheric Sciences, 25(6), 1-16. [2] Wang, G., J. Huang*, W. Guo, J. Zuo, J. Wang, J. Bi, Z. Huang, and J. Shi, 2010: Observation analysis of land-atmosphere interactions over the Loess Plateau of northwest China, J. Geophys. Res., 115, D00K17,doi:10.1029/2009JD013372. [3] Xie J., J. Huang*, G. Wang, K. Higuchi, J. Bi, Y. Sun, H. Yu, and T. Wang, 2010: The effects of clouds and aerosols on net ecosystem CO2 exchange over semi-arid Loess Plateau of Northwest China, Atmos. Chem. Phys., 10, 8205-8218.

Answer: We thank the reviewer for several helpful suggestions and we implemented as suggested.

[1] Huang, J.*, W. Zhang, J. Zuo, J. Bi, J. Shi, X. Wang, Z. Chang, Z. Huang, S. Yang, B. Zhang, G. Wang, G. Feng, J. Yuan, L. Zhang, H. Zuo, S. Wang, C. Fu and J. Chou, 2008: An overview of the Semi-Arid Climate and Environment Research Observatory over the Loess Plateau, Advances in Atmospheric Sciences, 25(6), 1-16.

[2] Wang, G., J. Huang*, W. Guo, J. Zuo, J. Wang, J. Bi, Z. Huang, and J. Shi, 2010: Observation analysis of land-atmosphere interactions over the Loess Plateau of northwest China, J. Geophys. Res., 115, D00K17,doi:10.1029/2009JD013372.

[3] Xie J., J. Huang*, G. Wang, K. Higuchi, J. Bi, Y. Sun, H. Yu, and T. Wang, 2010: The effects of clouds and aerosols on net ecosystem CO2 exchange over semi-arid Loess Plateau of Northwest China, Atmos. Chem. Phys., 10, 8205-8218.

Referee#2

General comments:

This ambitious study considers a range of mechanisms affecting CO_2 and CH_4 concentrations over the course of a year. Given the range of mechanisms involved, the approach needs to be far more systematic and the analysis more robust. In many places, the discussion does not relate closely enough to the data – results are presented and then an explanation is suggested based on literature, without any testing or demonstration of its relevance to this dataset. It is often not clear to the reader why a particular plot or grouping of data has been chosen. As many of the findings are not clear-cut, this leads the reader to question whether the results are robust or whether the conclusions would be different if data had been analysed slightly differently. In some places, there seems to be a very large jump between the data presented and the conclusions drawn. One key issue is the relative importance of the various mechanisms considered. In each subsection (of Section 4) the mechanism under consideration is used to explain the results as presented in that subsection, while the other processes (some of which have been shown to be major controls) are generally ignored.

In terms of the methodology, important information is missing about the study area in particular. It is often unclear how data have been averaged and why. The paper needs restructuring so that the reader understands the aims, approach and decisions taken by the authors.

The manuscript also has several typographical errors and language issues (not all detailed here).

Specific comments referring to particular lines are given below:

Introduction:

The Introduction needs restructuring and developing. A clear outline of objectives is needed. A summary of the various mechanisms that will be examined in the rest of the paper would improve readability. Previous work that is relevant to this study should be discussed.

Pg. 34207 Line 12-5: It is not clear why this sentence appears here. It would fit more naturally in Section 3.2.2.

Answer: We have incorporated in section 3.2.2.

Pg. 34207 Line 16-23: This meaning of this paragraph is unclear.

Answer: Revised as suggested in the updated manuscript.

Study area:

More information about the study area is required. Figure 1a is not very informative and the scale is difficult to read. An aerial image, map or photograph of the study area would be far more helpful. What is the land use and land cover? Please provide some information about the characteristics of buildings and/or vegetation. Please provide some context for this study compared to other similar studies. How large is the study area?

Answer: More details have been shown from figure 1a. The land use and land cover information added in the revised manuscript.

Pg. 34208 Line 3: The site is described as 'rural' here but 'suburban' in the title.

Answer: Corrected in the updated manuscript.

Pg. 34208 Line 4: Population density would be more useful to facilitate comparison with other sites.

Answer: Population density information provided in the revised manuscript.

5). Pg. 34208 Line 9: What is meant by 'near'? Please quantify.

Answer: Updated in the revised manuscript.

Data set and methodology:

Pg. 34208 Line 23-5: This paragraph does not communicate very much. It may be more informative to provide a brief summary of which variables are being measured or modelled and why here, before moving on to the subsections giving the details of each. Currently, the reader does not have a clear overview of the campaign.

Answer: We already presented in table.1.

In-situ observations:

More details are required about the experimental setup. Where are the sensors located (in terms of their surroundings and measurement height)?

Answer: Updated in the revised manuscript.

It is not clear how the data have been averaged. What is meant by 'diurnally averaged' (Pg. 34209 Line 19)? What temporal resolution was used in Fig 2? In Fig 2a-b are these monthly averages and variation of daily values or hourly values or something else? What about in Fig 2c-d? What do the error bars represent?

In general, more detail is needed in the figure captions.

Answer: Hourly data have been averaged. Fig 2a-b is updated as daily, weekly and monthly averages in the revised manuscript as reviewer suggested. Fig 2c-d updated as Fig 5a-d with additional boundary layer information as per referee suggestion. In the revised manuscript, Fig2c-d is monthly variation of GHGs against NDVI.

In Fig 1b-e are the data monthly averages? Indicate the data are for 2014.

Answer: Yes monthly, updated in the manuscript.

Please also put y-axis ticks at more intuitive intervals on all plots (e.g. 50, 60, 70, and 80% in Fig 1d)."

Answer: Corrected as suggested.

In Fig 3 and 4 what does each point represent? How have the data been averaged?

Answer: Daily average. In the current manuscript, figures have been updated with new numbering as per referee suggestion.

Results and discussion – the presentation and analysis of results needs significant improvement throughout this section. The discussion is often unclear and does not fully address the trends seen in the results. The explanations are often vague and, although processes are mentioned, they are not convincingly linked to the results of this study. The references used should be expanded here if relevant to this dataset, or used in the Introduction if they are useful as background instead.

Answer: More explanation with new references have been updated in the revised manuscript.

Seasonal variations:

12). Monthly averages are presented in Fig 2a-b but results are discussed in terms of seasons (consisting of 2, 3 or 4 months according to Section 2). Note it may be helpful to indicate the different seasons on Fig 2a-b.

Answer: Section 4.1 is updated with more explanation and new figure.

13). For CO_2, the seasonal averages are very similar to each other, so it does not make sense to provide seasonal values and then talk about differences between behaviour in each season. Fig 2a suggests there may be relatively high CO_2 near the start of the monsoon season, although the period of missing data and considerable variability means the picture is not especially clear. If 1-week or 2-week averages were used in Fig 2a instead, is the overall result the same? If the data are grouped according to the actual onset of the monsoon (rather than monthly approximations), are the results any more conclusive? The discussion and explanation (Pg. 34212 Line 4-13) does not give a clear overview of the processes involved, how they impact the CO2 concentration and when or why each process is most significant.

Answer: With more explanation, section 4.1 is updated in the revised manuscript.

14). Pg. 34212 Line 4 ‘loss of carbon’ from what?

Answer: Less carbon budget in winter due to respiratory losses (Aurela et al. 2004). Updated in the revised manuscript.

15). For CH_4, again, consideration should be given to the robustness and suitability of using monthly/seasonal averages. The analysis is vague and does not adequately explain the results. In particular, ‘associated with the Kharif season’ (Line 20) is vague and needs further explanation. Is the rate of change really highest during post-monsoon (OND) and winter (JF) (Line 26-7)? The final sentence in Section 4.1 does not explain the results; please state and explain precisely what is meant (rather than ‘This may be: : :’).

Answer: Present study analysis observed highest CH_4 concentration during post-monsoon and started decrease in subsequent seasons.

Diurnal variations:

16). Pg. 34213 Line 5-7: The meaning of this sentence is unclear. Could you provide an example specific to this dataset?

Answer: Sentence has been withdrawn and updated accordingly.

17). Pg. 34213 Line 14-6: Referring to other studies is helpful, but are the sites in those studies similar, i.e. are the same processes relevant? More detailed discussion needed.

Answer: Similar observations were made by Sharma et al. 2014 at Gadanki which has similar land use land cover as Shadnagar. Published results have been cited

18). Pg. 34213 Line 16-20: Needs more explanation. Do boundary layer dynamics affect CH4 concentrations as well? How does consideration of boundary layer height impact the findings from the previous subsection?

Answer: Diurnal variations of atmospheric species such as CO_2 and CH_4 mainly controlled by boundary layer dynamics. However, the source and sink mechanisms for these gases may be different. More explanation provided at section 4.4 and 4.5.1 in the revised manuscript.

Influence of prevailing meteorology:

19). Fig 3 – what does each point represent? Pg 34214 Line 2 mentions 'monthly mean wind speed'. Daily or hourly averages may be most suitable, bearing in mind the diurnal cycles seen in Fig 2.

Answer: Daily averages

20). Pg. 34214 Line 8-15: Wind direction and source area seem to be a very relevant consideration and should be addressed in more detail (again a map and some quantitative information would be useful).

Answer: Land use land cover information given in Fig1 and quantitative information on influence of wind direction provided in table 3.

21). Pg. 34214 Line 16 - Pg. 34215 Line 6: It is very difficult to relate the correlations discussed here to Figure 4, which leaves the reader rather unconvinced of the results. The analysis presented here does not seem sufficient to draw the conclusions reached in this section. Where other studies are used to try to explain potentially relevant processes, they are linked too vaguely to the results and there seems to be little evidence that these processes are actually relevant to the data shown here.

Answer: We tried to improve the quality of the figure for better visualization and interpretation. Meteorological processes which influence seasonal variations of GHGs has been provided in the updated manuscript.

22). Pg. 34214 Line 20: Is there any diurnal cycle in wind speed that should be accounted for? Do the findings change significantly if daily/hourly averages are used for wind speed/temperature/humidity?

Answer: These parameters (wind speed/temperature/humidity) will have diurnal behaviours as GHGs. We have not seen any significant between daily and hourly averages.

Influence of boundary layer height on GHGs mixing ratios:

23). the figure, discussion and conclusion do not give a clear picture of how the boundary layer height influences the mixing ratios.

Answer: X axis represents the seasonal transition i.e. monsoon to post monsoon (M-PM) etc. and y axis indicates seasonal difference of BLH and GHGs concentration respectively. We tried to bring out seasonal variation of BLH on GHGs using satellite and diurnal effect from ECMWF-ERA data sets.

Methane sinks mechanism:

24). Most of Section 4.5 would be better in the Introduction (which would also help the reader in Section 3 when the various datasets are described). Might the high CH4 readings be due to the highway and railway directly? The dependence on OH seems like a hypothesis which can be neither supported nor rejected based on the analysis presented here.

Answer: For the continuity of manuscript, we updated section 4.5 as 4.6 in the revised manuscript. Yes, we observed high NOx values from the eastern direction (where highway and railways are there) which subsequently decreases OH radical through chemical process as described in the section 4.6. Due to which high values of CH_4 were observed during case study period.

Influence of vegetation:

25). Pg. 34218 Line 22-3: 'NDVI showed inverse relationship with CO2, mainly due to change in vegetation which affects the CO2 concentrations.' Both halves of this sentence effectively say the same thing without explaining the process.

Answer: Typo error, sentence is reconstructed and updated in revised manuscript.

To summarise, Section 4 contains too many different mechanisms without consideration of how they impact each other or a clear systematic structure to the analysis. Perhaps the relationship with NDVI should be moved closer to the start of the section where seasonal variations are discussed. Looking at the monthly ratio (Fig 6) may also be more useful earlier on. The high CH4 readings in Fig 7 may need to be discussed alongside source area analysis in Section 4.3.

Are there other sources or sinks of GHGs which have not been considered in this analysis? How might they impact the results?

Answer: As per your suggestions, manuscript is rearranged and updated with more explanation.

Conclusions:

26). the conclusion should draw together the findings and provide an insightful summary of the research. Many statements are vague (e.g. Pg 34221 Line 7-8: 'This clearly indicates the seasonal variations in source-sink mechanisms of CO2 and CH4 respectively.' What are the source-sink mechanisms and how do they differ for CO_2 and CH_4 with season?) What new findings have emerged from analysis of this dataset?

Answer: As per your suggestions, manuscript is updated.

Minor comments:

1). Throughout: Change GHG's to GHGs

Answer: Updated in the revised manuscript.

2). Pg 34206 Line 11-2: Sentence not clear – please rephrase.

Answer: Rephrased as suggested

3). Pg 34206 Line 16-7: Sentence vague and not clear– please rephrase.

Answer: Updated in the revised manuscript.

4). In many places spaces are missing, e.g. Pg. 34206 Line 22: '(GHG),particularly'; Pg. 34206 Line 24: 'emissionsand'; Pg 34207 Line 8: 'andecosystems' ; Pg 34207 Line 9: 'reflector'

Answer: Updated in the revised manuscript.

5). Pg 34206 Line 26: What is the significance of May 2013? A longer-term perspective that extends to the present may be more useful (i.e. to indicate May 2013 is not an exception).

Answer: This sentence has been modified in the revised manuscript.

6). Pg 34209 Line 3: I would mention 'Los Gatos Research' here rather than in the Abstract and Introduction.

Answer: Updated in the revised manuscript.

7).Pg. 34209 Line 5-7: Reference would be useful here.

Answer: Updated with Berman et al., 2012; Shea et al., 2013: Mahesh et al., 2015.

8). Pg. 34209 Line 20-1: Give exact dates.

Answer: We included exact dates in the revised manuscript.

9). Pg. 34209 Line 25: Correct brackets.

Answer: The correction is incorporated in the revised manuscript.

10). Pg. 34211 Line 24: Better to define mean and standard deviation here rather than in the Abstract.

Answer: This has been changed in the revised manuscript.

11) Pg. 34212 Line 3-4: Change to ': : : ppm in winter, pre-monsoon, monsoon and postmonsoon, respectively'

Answer: Updated in the revised manuscript.

12). Pg. 34213 Line 22: Change 'place' to 'plays'

Answer: Updated in the revised manuscript.

Influence of Meteorology and interrelationship with greenhouse gases (CO_2 and CH_4) at a sub-urban site of India

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Abstract

Atmospheric greenhouse gases (GHGs), such as carbon dioxide (CO_2) and methane (CH_4), are important climate forcing agents due to their significant impacts on the climate system. 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. The annual mean ~~of~~ CO_2 and CH_4 over the study region are~~is~~ found to be 394±2.92 ppm and 1.92±0.07 ppm ~~(mean~~ (μ~~)~~ ±1~~std (~~σ)~~)~~ respectively. CO_2 and CH_4 show~~ed~~ a significant seasonal variation during the study period with maximum (minimum) CO_2 observed during Pre-monsoon (Monsoon), while CH_4 recorded maximum during post-monsoon and minimum in monsoon. Irrespective of the seasons, consistent diurnal variations of ~~A consistent diurnal mixing ratio of~~ these gases are ~~is~~ observed. ~~with high (low) during night (afternoon) hours throughout the study period.~~ Influences of prevailing meteorology (air temperature, wind speed, wind direction and relative humidity) on GHG~~'~~s have also been investigated. CO_2 and CH_4 show~~ed~~ a strong positive correlation during winter, pre-monsoon, monsoon, and post-monsoon with correlation coefficients (Rs)~~R~~ equal to 0.80, 0.80, 0.61, and 0.72 respectively; indicating common anthropogenic source for these gases.~~. It implies the seasonal variations in source-sink mechanisms of CO_2 and CH_4.~~ Analysis of this study reveals the major sources for CO_2 are soil respiration and anthropogenic emissions while vegetation act as a main sink. Whereas the major source and sink for CH_4 are vegetation and presence of hydroxyl (OH) radicals.

Keywords: Carbon dioxide, Methane, OH radical.

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1. Introduction

The Intergovernmental Panel on Climate Change (IPCC, 2013) reported that humankind is causing global warming through the emission of greenhouse gases (GHGs), particularly carbon dioxide (CO_2) and methane (CH_4).CO_2 and CH_4 concentrations have increased by 40% and 150 % respectively since pre-industrial times, mainly from fossil fuel emissions and secondarily from net land use change emissions (IPCC, 2013; Huang et al., 2015). CO_2 measurements at MaunaLoa, Hawaii (Monastersky, 2013) have exceeded the 400 ppm mark several times in May 2013. CH_4 is also receiving increasing attention due to high uncertainty in its sources and sinks (Keppler et al., 2006; Miller et al., 2007; Frankenberg et al., 2008). Stefanie 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 (A. Garg et al., 2001; StefanieKirschke et al., 2013). In India, electric power generation that contributes to half of India's total CO_2 equivalent emissions (A. Garg et al., 2001).

Arid and semi-arid areas comprise about 30% of the Earth's land surface. Climate change and climate variability will likely have a significant impact on these regions (Huang et al., 2008; Huang et al., 2015). The variability of environmental factors may result in significant effects on regional climate and global climate (Wang et al., 2010), especially the radiative forcing; via the biogeochemical pathways affecting the terrestrial carbon cycle. 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 (Liu et al., 2011). The planetary boundary layer (PBL) is the part of the atmosphere closest to the Earth's surface where turbulent processes often dominatethe 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 (OḢ) 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 ruralsuburban location situated ~70km away from urban site of Hyderabad (Northern side) with a population of ~0.16 million (Patil et al., 2013). A schematic map of study area is shown in Fig. 1a. Major sources 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, Shadnagar Campus (17°02'N, 78°11'E). Sampling site is near (aerial distance ~ 2.25 km) to National highway 7 (NH7) and a railway track (non-electrified) is in the East (E) direction.

Mean monthly variations of temperature (°C) and relative humidity (RH (%) observed at Shadnagar during 2014 are shown in Figure 1e and 1d 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°c 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. 1c) varies between 1.3 to 1.6 m s^{-1} with a maximum observed during monsoon and minimum in pre-monsoon. The air mass advecting (Fig. 1b) 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.

3.1 In-situ observations

3.1.1 Greenhouse Gas Analyser (GGA)

The Los Gatos Research's - Greenhouse Gas Analyser (model: LGR-GGA-24EP) is an advanced instrument capable of simultaneous measurements of CO_2, CH_4 and H_2O. This instrument is well known for high precision and accuracy which are crucial towards understanding background concentrations of atmospheric GHGs, with specifications meeting WMO standards of measurement (Berman et al., 2012; Shea et al., 2013: Mahesh et al., 2015). It is based on enhanced Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS) technology (Paul et al. 2001, Baer et al., 2002), which utilizes true wavelength scanning to record fully resolved absorption line shapes. Considering the ~~rural~~ nature of the site, flow rate is fixed to be 7 liters per minute (lpm). Ambient air entering the GGA is analysed using two near infrared (NIR) distributed feedback tunable diode lasers (TDL), one for a CO_2 absorption line near1.60 µm (v_0= 6250 cm^{-1}) and the other to probe CH_4 and H_2O absorption lines near 1.65 µm (v_0= 6060.60 cm^{-1}). The concentration of the gases is determined by the absorption of their respective characteristic absorption lines with a high sampling time of 1sec. A detailed explanation regarding the configuration, working and calibration procedure performed for GGA in NRSC can be found elsewhere in Mahesh et al., (2015). In the present study we used GGA retrieve~~d~~ CO_2 and CH_4 data. High resolution data sets are diurnally averaged and ~~is~~ used in further analysis. Due to failure of internal central processing unit (CPU) of the analyzer, data are~~is~~ not recorded from pre-monsoon month of 1st May to ~~a few days in~~ 18th June during the study period.

3.1.2 O_3 and NO_x analyzer

Surface concentrations of O_3 and NO_X have been measured continuously using on-line analyzers (Model No.s: 49iand 42i for O_3 and NO_X respectively), procured from Thermo Scientific, USA~~)~~ since July 2014. The trace gases (O_3 and NOx) sampling inlet is installed on the top of a 2 m mast fixed on the roof of an 8 m high building, and ambient air flow is supplied to the instruments. The inlet prevents the ingress of rain water, and is equipped with 0.5 µm filter to prevent accumulation of dust within the instrument. The ozone analyzer is based on Beer-Lambert-

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Baugher law which relates absorption of light to the concentration of species as its 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 NO_2 into NO and estimates the NO_x concentration by the intensity of light emitted during the chemiluminescent reaction of NO with O_3 present in the ambient air. The analyzer is integrated with zero and span calibrations which are performed twice monthly.

Simultaneous observations of meteorological parameters are obtained from an automatic weather station~~s~~ (AWS) ~~located in the same campus.~~ installed in NRSC, Shadnagar campus as a part of Calibration and Validation (CAL/VAL) project in March 2012 is equipped with nine sensors to measure fifteen weather parameters. Weather parameters measured are at surface level and height of the AWS mast is ~10 meters. ~~Wind speed and direction measurements are collected at the maximum height (3m) and all others are at 1-1.5m height.~~

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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 m, 500 m, and 1 km (channels 8–36), covering the visible, near-infrared, shortwave 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:

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

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).

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 microsatellites, 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 (such as, temperature and humidity profiles)~~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). The planetary boundary layer (PBL) is 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).

3.2.3 Hysplit model

The general air mass pathway reaching over Shadnagar is analysed using HYSPLIT model (Draxler and Rolph, 2003) [http://ww.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 00:60 UTC and reaching study site, (Shadnagar) at different altitudes(1 km,2 km,3km 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 CO_2 and CH_4

Temporal~~Monthly~~ variations of CO_2 and CH_4 during the study period are shown in Figure. 2a and 2b. The circles indicate the daily mean, while triangular markers represent weakly averages and monthly mean by square markers. ~~-~~Annual mean of CO_2 over study region is found to be 394±2.92 (mean (μ) ± standard deviation (1σ~~)~~)~~(μ ± 1σ)~~) ppm with an observed minimum in monsoon and maximum in pre-monsoon. Seasonal mean~~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 in~~with respectively~~ winter, pre-monsoon, monsoon, and post-monsoon respectively. Minimum CO_2 during winter (dry season) can be due to respiratory~~indicates the~~ loss of carbon (Gilmanov et al., 2004; Aurela et al. 2004) as decreased temperature and solar radiation during this period inhibit increases in local CO_2 assimilation (Thum et al., 2009). A steady increase in CO_2 concentration is observed as season changes from winter to pre-monsoon months. 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). The enhanced soil respiration during these months also compliments the increase in CO_2 concentration during this period. In addition to these natural causes, biomass burning over Indian region can also have a significant effect on pre-monsoon CO_2 concentration. More detailed explanation of biomass burning influence on pre-monsoon GHGs concentration is discussed in section 4.6.- 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. A decrease in CO_2 concentration is also observed as the monsoon progress. The decreases in temperature (due to cloudy and overcast conditions prevailing during these months) reduce leaf and soil respiration which contributes to the enhancement of carbon uptake (Patil et al., 2013; Jing et al., 2010). Further increase during post-monsoon CO_2 is associated with high ecosystem productivity (Sharma et al., 2014) also an enhancement in soil microbial activity (Stefanie Kirschke et al., 2013).

CH_4 concentration in the troposphere is principally determined by a balance between surface emission and destruction by hydroxyl radicals ($\dot{OH}$). 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. Seasonal mean (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. The highest concentration appears during post-monsoon and may be associated with the Kharif season (Goroshiet al., 2011). Seasonal meanBackground (average) values of CH_4 observed during different seasons are 1.93±0.05, 1.89±0.05, and 1.85±0.03, and 2.02±7 ppm with respectively winter, pre-monsoon, monsoon, and post-monsoon. Hayashida et al. (2013) reported that Tthe seasonality of CH_4 concentration over monsoon Asia is characterized by higher values in the wet season and lower values in the dry season; possibly because of the effects of strong emissions from rice paddies and wetlands during the wet season. 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 (Abhishek 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 (Nishanth et al., 2014; Goroshiet al., 2011). Due to this, This may be one of the major reasons for the enhanced CH_4 observed during post-monsoon at present study area.and winter seasons (Nishanth et al., 2014; Sheshakumar et al., 2011).

4.2 Influence of vegetation on GHGs.

In India cropping season is classified into (i) Kharif and (ii) Rabi based on the onset of monsoon. The Kharif season is from July to October during the south-west (SW) monsoon and Rabi season is from October to March (Koshal Avadhesh, 2013). NDVI being one of the indicators of vegetation change, monthly variations of CO_2 and CH_4 against NDVI is studied to understand the impact of land use land cover on mixing ratios of CO_2 and CH_4. Monthly mean changes in NDVI, CO_2 and CH_4 are shown in Figure 2c and 2d. Monthly mean of GHGs represented in this analysis is calculated from daily mean in day time (10-16 LT). Analysis of the figure reveals that an inverse relationship exists between NDVI and CO_2; while a positive relation is observed w.r.t CH_4. Generally over this part of the country vegetation starts during the month of June with the onset of SW monsoon and as vegetation increases a decrease in CO_2 concentration is observed, due to enhancement in photosynthesis. Further a decline in NDVI is observed as the season advances from post monsoon to winter and then to pre-monsoon, and it is associated with an increase in CO_2 concentration. Similarly, -the main source for CH_4 emissions are soil microbial (Stefanie Kirschke et al., 2013) activity which are more active during monsoon and post monsoon seasons. High (low) soil moisture and NDVI is observed in monsoon (pre-monsoon) seasons (Figure 8a and 8b). The predominating factors which controls the soil emissions of CO_2, CH_4 are moisture content, soil temperature, vegetation and soil respiration (Smith et al., 2003; Jones et al., 2005; Chen et al., 2010) respectively.

Biomass burning (forest fire and crop residue burning) is one of the major sources of gaseous pollutants such as carbon monoxide (CO_2), methane (CH_4), nitrous oxides (NOx) and hydrocarbons in the troposphere (Crutzenet al., 1990, 1985; Sharma et al., 2010). In order to study the role of biomass burning on GHGs a case study is discussed. Figure 43c shows the spatial distribution of MODIS derived fire counts over Indian region during 14-21 April 2014 with air mass trajectories ending over study area over layed on it at different altitudes viz. 1000 m, 2000 m and 4000 m respectively. Analysis of the figure shows a number of potential fire

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locations on the north-western and south-eastern side of study location and trajectories indicate its possible transport to study area. Daily mean variation of GHGs during the month of Aril 2014 (Figure 43b) indicates an enhancement in GHGs during the same period (14-21 April 2014). Analysis reveals that CO_2 and CH_4 have increased by ~2% and ~0.06% respectively during event days with respect to monthly mean. This analysis reveals that long range / regional transported biomass burning have a role in enhancement of GHGs over study site. Further to understand the seasonal variation of biomass burning contribution to GHGs we analysed long term (2003-2013) Fire Energetics and Emissions Research version 1.0 (FEER v1) data over study area. Emission coefficient (C_e) products during biomass burring is developed from coincident measurements of fire radiative power (FRP) and AOD from MODIS Aqua and Terra satellites (Ichoku and Ellison, 2014). Figure 43a shows seasonal variation of CO_2 emission due to biomass burning over the study site. Enhancement in CO_2 emission is seen during pre-monsoon months; which also supports earlier observation (Figure 2a). This analysis reveals that biomass burning has a role in pre-monsoon enhancement of CO_2 over study site. For a qualitative analysis of this long range transport, we have analysed air mass trajectories ending over study site during different seasons.

4.3 *Correlation between CO_2 and CH_4*

A correlation study is carried out between hourly averaged CO_2 and CH_4 during all season for the entire study period. The statistical analysis for different seasons is shown in Table 32. Fang et al., (2015) suggest the correlation coefficients (Rs) value higher than 0.50 indicates a similar source mechanism of CO_2 and CH_4. Also a positive correlation dominance of anthropogenic emission on carbon cycle. Our study also reveals a strong positive correlation observed between CO_2 and CH_4 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}, ψ_{y-int}) are computed using Taylor (1997) which showed maximum during winter, pre-monsoon, and minimum in a monsoon that figure out the hourly stability of the mixing ratios between CO_2 and CH_4. This can be due to relatively simple source/sink process of CO_2 in comparison with CH_4. Figure 54 shows the seasonal variation of $\Delta CH_4/\Delta CO_2$. Dilution effects during transport of CH_4 and CO_2 can be minimized to some extent by dividing the increase of CH_4 over time by the respective increase in CO_2 (Worthy et al., 2009). In this study, background concentrations of respective GHGs 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 $\Delta CH_4/\Delta CO_2$ over the study region during the study period is found to be 7.1 (ppb/ppm). This low value clearly indicates the dominance of CO_2 over the study region. The reported $\Delta CH_4/\Delta CO_2$ values from some of the rural sites viz Canadian Arctic and Hateruma Island (China) are of the order 12.2 and ~10 ppb/ppm respectively (Worthy et al., 2009; Tohjima et al., 2014). Average $\Delta CH_4/\Delta CO_2$ ratio during winter, pre-monsoon, monsoon and post-monsoon are 9.40, 6.40, 4.40, and 8.20 ppb respectively. Monthly average, of $\Delta CH_4/\Delta CO_2$, is relatively high from late post-monsoon to winter, when the biotic activity is relatively dormant (Tohjima et al., 2014). During pre-monsoon decease in $\Delta CH_4/\Delta CO_2$ ratio indicates the enhancement of CO_2 relative to that of CH_4.

4.24.4 Diurnal variations of CO_2 and CH_4

Figure 25ea to 5d and 2d shows the seasonally averaged diurnal cycle of CO_2 and CH_4 over Shadnagar during study period. The vertically bar represents the standard deviation from respective mean. Irrespective of seasonal variation GHGs showed a similar diurnal variation, with maximum mixing ratios observed during early morning (06:00 hrs) as well as early night hours (20:00 hrs) and minimum during afternoon hours. Figure 2c and 2d depicts seasonal diurnal variations of CO_2 and CH_4 over Shadnagar during study period. The amplitudes diurnal changes during seasonal variation mainly depend on biosphere sources and sinks such as land use and land cover change (Fearnside 2000, IPCC, and AR5). Maximum mixing ratios of CO_2 and CH_4 are observed during early morning and late night hours. Peak surface concentrations of CO_2 and CH_4 increase at night and remain high until sunrise (22:00hrs to 06:00hrs). However the difference observed in the maximum diurnal amplitudes can be attributed to seasonal changes. The observed diurnal cycle of GHGs is closely associated with diurnal variation of planetary boundary layer height (PBLH). For better understanding of the diurnal behavior of CO_2/CH_4, we used European Centre for Medium-range Weather Forecasting (ECMWF) Interim Reanalysis (ERA) PBL data set which gives the data for every three hours viz. 00:00, 03:00, 06:00, 09:00, 12:00, 15:00, 18:00, and 21:00 UTC with a resolution of 0.25°x0.25° (http://data-portal.ecmwf.int). Figure 5a to 5d portrays the diurnal evolution of CO_2/CH_4 during different season along with the evolution of Boundary Layer Height (m) on secondary y axis. The morning peak arises due to combined

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influence of fumigation effect, (Stull 1988) and morning build-up of local anthropogenic activities (household and vehicular transport). Low value of GHGs as the day progress can be attributed to increased photosynthetic activity during day time and destruction of stable boundary layer and residual layer due convective activity. In the evening hours, surface inversion begins and form a shallow stable boundary layer (Nair et al., 2007) causing the enhancement in GHGs concentration near the surface. ~~Figure 2c shows mixing ratios of CO_2 are gradually decreasing after sun rise and reaching peak minimum in the afternoon because of the net ecosystem uptake of the biosphere and boundary layer dynamics. During night time, mixing ratios increase due to formation of stable atmospheric boundary layer, soil respiration of the biosphere and absence of photosynthetic activity.~~ Similar trend in diurnal variation of GHG~~'~~s is reported from other parts of the country (Patil et al., 2013; Mahesh et al., 2014; Sharma et al., 2014; Nishanth et al., 2014). ~~Although diurnal variations of CH_4 showed similar trend as of CO_2, but are caused due to different factors. Lower troposphere acts as main sink for CH_4 with the formation of O_3 through oxidation of CH_4 and other trace species in the presence of NO_x and hydroxyl radicals ($\dot{O}H$) (Eisele et al., 1997, IPCC, AR5).~~

~~4.3~~4.5 Influence of prevailing meteorology

Redistribution (both horizontal and vertical) of GHG~~'~~s also plays~~place~~ a role in their seasonal variation, as it controls transport and diffusion of pollutants from one place to another (Hassan 2015). A good inverse correlation between wind speed and GHG~~'~~s suggest the proximity of sources near measurement site, while a not so significant correlation suggests the influence of regional transport (Ramachandran and Rajesh, 2007). Figure ~~3~~6a and ~~3~~6b shows scatter plot between GHG~~'~~s and wind speed during different seasons. Analysis of Figure ~~3~~6 shows that there exists an inverse correlation between daily~~monthly~~ mean wind speed and GHG~~'~~s. Correlation coefficients (Rs) between wind speed and CO_2 during pre-monsoon, monsoon, post-monsoon, and winter is 0.56, 0.32, 0.06, and 0.67 respectively. While for CH_4 it is found be 0.28, 0.71, 0.21, and 0.60 respectively. Negative correlation indicates that the influence of local sources on GHG~~'~~s, however, poor correlation coefficients during different seasons suggest the role of regional/local transport (Mahesh et al 2014). Also an understanding of prevailing wind direction and its relationship with GHG~~'~~s helps in determining their probable source regions. Table ~~2~~3 shows the monthly mean variation of CO_2 and CH_4 with respect to different wind direction. Enhancement in CO_2 and CH_4 level over Shadnagar are observed to mainly come from NW and NE while the lowest is from the S and SW. This can be associated to some extend with industrial emissions located in western side of sampling site, and the influence of emission and transport from nearby urban center on the NW side of the study site.

The influence of meteorological parameters (temperature and relative humidity) influenceon trace gases is also examined. Figure 47a and 7b (top panel corresponds to CO_2 and bottom panel represents CH_4) shows the scatter plot of temperature versusvs. relative humidity as a function of GHGs during different seasons. Here, dDaily mean data is used instead of hourly mean data, to avoid the influence of the diurnal variations on correlations. CO_2 showed a positive correlation with temperature during all season except during winter. This negative correlation can be attributed to different response of photosynthesis rate to different air temperaturedecrease in rate of photosynthesis. IPCC (1990) reports that many mid-latitude plants shows an optimum gross photosynthesis rate when temperature varied from of 20 to 35 °C. The rate of plant respiration tends to be slow below 20°C. However, at higher temperatures, the respiration rate accelerates rapidly up to a temperature at which, it equals the rate of gross photosynthesis and there can be no net assimilation of carbon. While CH_4 showed a weak positive correlation with temperature during pre-monsoon and post-monsoon, while a weak negative correlation is observed during monsoon and winter. This could be due to the rate of chemical loss reaction with OH is faster in summer and minimum in other seasons. A case study on CH_4 sink mechanism has discussed in section 4.6. This indicates that regional air temperature doesn't 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 (Abhishek et al., 2014).

Figure 8a and 8b illustrates the daily mean variation of GHGs with respect to soil moisture and soil temperature (Top panel represent the seasonal variation of CO_2 w.r.t soil moisture and soil temperature, while bottom panel represent the seasonal variation of CH_4 against the same parameterswith the same). It's quite interesting to observe that GHGs behave differently w.r.t soil moisture during different seasons. CH_4 shows a positive relationship during monsoon and post-monsoon and an inverse relationship exist during pre-monsoon and winter; while a reverse

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relationship exist for CO_2. During wet season aeration is restricted (Smith et al. 2003) hence soil respiration is limited, which decrease CO_2 flux. This can be one of the factors for low values of CO_2 during monsoon months, during dry months soil may act as sink of CH_4.

~~4.3.1~~4.5.1 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 and it ~~.~~ closely resembles with the air temperature pattern. The highest (lowest) BLH over study region was identified 3.20 km (1.50 km). A~~n~~ monthly average ~~monthly~~ air temperature is maximum (minimum) of 29°C (20°C) during the summer (winter) months.

Seasonal BLH during winter, pre-monsoon, monsoon and post monsoon are 2.10 km, 3.15 km, 1.74 km and 2.30 km respectively.~~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); i~~Its influence on CO_2 and CH_4 mixing ratios are shown in Figure ~~5~~9a and ~~5~~9b. X axis represents the seasonal transition i.e. monsoon to post monsoon (M-PM) etc and y axis indicates seasonal difference of BLH and GHGs concentration respectively. As seasonal BLH ~~thickness~~ increase, mixing ratios of CO_2 (CH_4) decreased from 8.68 ppm to 5.86 ppm (110 ppb to 40 ppb). This effect clearly captured by seasonal diurnal averaged BLH data sets used from ECMWF-ERA. 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~~BLH ~~thickness~~ through convection and biosphere activities.

~~*4.4 Correlation between CO_2 and CH_4*~~

~~A correlation study is carried out between hourly averaged CO_2 and CH_4 during all season for the entire study period. The statistical analysis for different seasons is shown in Table 3. Fang et al., (2015) suggest the correlation coefficient (R) value higher than 0.50 indicates a similar source mechanism of CO_2 and CH_4. Also a positive correlation dominance of anthropogenic emission on carbon cycle. Our study also reveals a strong positive correlation observed between CO_2 and CH_4 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 a monsoon that figure out the hourly stability of the mixing ratios between CO_2 and CH_4. This can be due to relatively simple source/sink process of CO_2 in comparison with CH_4. Dilution effects during transport of CH_4 and CO_2 can be minimized to some extent by dividing the increase of CH_4 over time by the respective increase in CO_2 (Worthy et al., 2009). Figure 6 shows the seasonal variation of $\Delta CH_4/\Delta CO_2$. In this study, background concentrations 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 $\Delta CH_4/\Delta CO_2$over the study region during the study period is found to be 7.1 (ppb/ppm). This low value clearly indicates the dominance of CO_2 over the study region. The reported $\Delta CH_4/\Delta CO_2$ values from some of the rural sites viz Canadian Arctic and Hateruma Island (China) is of the order 12.2 and ~10 ppb/ppm respectively (Worthy et al., 2009; Tohjima et al., 2014). Average $\Delta CH_4/\Delta CO_2$ ratio during winter, pre-monsoon, monsoon and post-monsoon are 9.40, 6.40, 4.40, and 8.20 ppb respectively. Monthly average, of $\Delta CH_4/\Delta CO_2$, is relatively high from late post-monsoon to winter, when the biotic activity is relatively dormant (Tohjima et al., 2014). During pre-monsoon decease in $\Delta CH_4/\Delta CO_2$ ratio indicates the enhancement of CO_2 relative to that of CH_4.~~

~~*4.5*~~*4.6* ***Methane (CH_4) sink mechanism***

Methane (CH_4) is the most powerful greenhouse gas after CO_2 in the atmosphere due to its strong positive radiative forcing (IPCC, AR5). Atmospheric CH_4 is mainly (70-80%) from biological origin produced in anoxic environments, by anaerobic digestion of organic matter (Crutzen and Zimmermann, 1991). The major CH_4 sink is oxidation by hydroxyl radicals (OH), which accounts for 90 % of CH_4 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 (O_3) and water vapor (H_2O). Eisele et al., (1997) defined primary and secondary source of OH radicals in the atmosphere. Primary source of OH radical is as follows;

$$\mathrm{O_3 + hv\ (\lambda \leq 310\ nm) \rightarrow O_2 + O(^1D)} - - - (2)$$

$$\text{where}\mathrm{O(^1D)}\text{ is electronically excited atom}$$

$$\mathrm{O(^1D) + O_2 \rightarrow O + M} - - - (3)$$

$$\mathrm{O(^1D) + H_2O \rightarrow 2OH} - - - (4)\quad \text{primary OH formation}$$

Removal of CH_4 is constrained by the presence of OH radicals in the atmosphere. A 1 min time series analysis of CH_4, NO_x, O_3 and H_2O and associated wind vector for August 2014 to understand the CH_4 chemistry is shown in Figure ~~7~~10a and Figure ~~7~~10b. Low NO_x (1-2 ppb) values are shown in horizontal elliptical region of Figure ~~7~~10a and observed corresponding low CH_4 (1.80 ppm) concentrations. The low NO_x in turn produces high OH radicals in the atmosphere due to conversion of HO_2 radical by NO, which removes CH_4 through oxidation process as shown below.

$$\mathrm{HO_2 + NO \rightarrow O\dot{H} + NO_2} - - - (5)\text{ when }\mathrm{NO_x}\text{ levels }1 - 2\text{ ppb}$$

$$\mathrm{CH_4 + O\dot{H} \rightarrow C\dot{H}_3 + H_2O} - - - (6)\quad \text{main }\mathrm{CH_4}\text{ removal process}$$

$$\mathrm{NO_2 + OH + M \rightarrow HNO_3 + M} - - - (7)\text{ if }\mathrm{NO_x} > 2\ ppb\ (OH \downarrow, CH_4 \uparrow)$$

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 HO_2 and peroxy radicals (RO_2) react with NO to form OH radicals. Therefore OH radicals are much higher in the case of low NO_x. When NO_x levels increase more than 2 ppb, most of the OH radicals react with NO_2 to form nitric acid (HNO_3). In first order, the levels of CH_4 in the atmosphere depend on the levels of NO_x though the production of OH radicals in the atmosphere is still uncertain. Figure ~~7~~10a and ~~7~~10b showed high CH_4, H_2O, O_3 and NO_x during a few days in August 2014. High concentrations of CH_4, NO_x and other gases are observed in the eastern direction of study site. Very high NO_x levels above 10 ppb are observed and subsequently CH_4 concentrations also increased to 2.40 ppm from 1.80 ppm. In the eastern direction of study site a national highway and single line broad gauge railway network are present which act as possible sources of NOx, CH_4 and CO_2. Increase in emissions of NO_x causes decline in the levels of OH radicals and subsequently observed high CH_4 over the study region.

4.6 *Influence of vegetation on GHG's.*

In India cropping season is classified into (i) Kharif and (ii) Rabi based on the onset of monsoon. The kharif Kharif season is from July to October during the south-west monsoon and Rabi season is from October to March (Avadhesh Koshal, 2013). NDVI being one of the indicators of vegetation change, monthly variations of CO_2 and CH_4 against NDVI is studied to understand the impact of land use land cover on mixing ratios of CO_2 and CH_4. Monthly changes in NDVI, CO_2 and CH_4 are shown in Figure 8. Monthly mean of GHG's represented in this analysis is calculated from daily day time (10-16 LT) mean. Maximum NDVI of 0.60 corresponding to the minimum CO_2 concentration (about 382 ppm) is observed in September. NDVI showed inverse relationship with CO_2, mainly due to change in vegetation which affects the CO_2 concentrations. Initially during the month of June vegetation start increasing with availability of water and as vegetation increases .concentration. Similarly, The main source for CH_4 emissions are soil microbial (Stefanie Kirschke et al., 2013) activity which are more active during monsoon and post monsoon seasons. High (low) soil moisture and NDVI is observed in monsoon (pre-monsoon) seasons (Figure 9a and b). The predominating factors which controls the soil emissions of CO_2, CH_4 are moisture content, soil temperature, vegetation and soil respiration (Smith et al., 2003; Jones et al., 2005; Chen et al., 2010) respectively.

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Biomass burning (forest fire and crop residue burning) is one of the major sources of gaseous pollutants such as carbon monoxide (CO), methane (CH4), nitrous oxides (NOx) and hydrocarbons in the troposphere (Crutzenet al., 1990, 1985; Sharma et al.,2010). In order to study the role of biomass burning on GHG's a case study is discussed. Figure 10a shows the spatial distribution of MODIS derived fire counts over Indian region during 14-21 April 2014 with air mass trajectories ending over study area over layed on it at different altitudes viz. 1000m, 2000m and 4000m respectively. Analysis of the figure shows a number of potential fire locations on the north-western and south-eastern side of study location and trajectories indicates its possible transport to study area. Daily mean variation of GHGs during the month of Aril 2014 (Figure

10b) indicates an enhancement in GHGs during the same period (14-21 April 2014). Analysis reveals that CO_2 and CH_4 has increased by ~2% and ~0.06% respectively during event days with respect to monthly mean. This analysis reveals that long range / regional transported biomass burning have a role in enhancement of GHGs over study site. Further to understand the seasonal variation of biomass burning contribution to GHGs we analysed over study site we have analysed GHG's emissions from biomass burning using long term (2003-2013) Fire Energetics and Emissions Research version 1.0 (FEER v1) data over study area. Emission coefficient (C_e) products during biomass burring is developed from coincident measurements of fire radiative power (FRP) and AOD from MODIS Aqua and Terra satellites (Ichoku and Ellison, 2014). Figure 10c shows seasonal variation of CO_2 emission due to biomass burning over the study site. Enhancement in CO_2 emission is seen during pre-monsoon months; which also supports earlier observation (Figure 2a). This analysis reveals that biomass burning has a role in pre-monsoon enhancement of CO_2 over study site. For a qualitative analysis of this long range transport, we have analysed air mass trajectories ending over study site during different seasons.

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4.7 Long range circulations

To understand the role of long range circulation, we separated the trajectory into 4 clusters based on their pathway, namely North-East (N-E), North-West (N-W), South-East (S-E), South-West (S-W). The main criterion of trajectory clustering is to minimize the variability among trajectories and maximize variability among clusters. Cluster mean trajectories of air mass and their percentage contribution to the total calculated for each season over the study period at 3 Km altitude are depicted in Figure 911. Majority of air mass trajectories during winter (~44%), pre-monsoon (~64%), monsoon (~80%) and post-monsoon (~41%) are originating from NW parts of the study site. For a comprehensive analysis, percentage occurrences of cluster mean trajectories of air mass over study area during different season at different altitudes are also tabulated in Table 4. During post-monsoon to early pre-monsoon periods which are generally the post-harvest period for some of the crops agriculture residue burning which are quite common in the NW and NE regions part of India (Sharma et al, 2010).Our analysis reveals that during this period majority of air mass reaching the study site at different altitudes come from this part of the country.

5. Conclusions

The present study analysed the seasonal variations of atmospheric GHG~~'~~s (CO_2 and CH_4) and associated prevailing meteorology over Shadnagar, a suburban site of Central India during the period 2014. The salient findings of the study are the following:

- Irrespective of seasons, major sources for CO_2 are soil respiration and anthropogenic emissions while vegetation acts as a main sink. Whereas the major source and sink for CH_4 are vegetation and presence of hydroxyl (OH) radicals. In addition, boundary layer dyanamics and long range transport also plays a vital role on GHGs mixing ratios.
- The annual mean of CO_2 and CH_4 over the study region are~~is~~ found to be 394±2.92 ppm and 1.92±0.07 ppm (μ±1σ) respectively. CO_2 and CH_4 show~~ed~~ a significant seasonal variation during the study period. Maximum (Minimum) CO_2 is observed during Pre-monsoon (Monsoon), while CH_4 recorded maximum during post-monsoon and minimum in monsoon. Seasonal analysis of FEER data also showed maximum emission of CO_2 due to biomass burning during pre-monsoon months which indicates the influence of biomass burning on local emissions.
- CO_2 and CH_4 showed consistent diurnal behavior in spite of their significant seasonal variations, with an observed morning (06:00 IST) maxima, followed by afternoon minima (14:00 IST) and enhancing in the late evening (~22:00 IST).
- Correlation coefficient (Rs) between wind speed and CO_2 during pre-monsoon, monsoon, post-monsoon and winter is 0.56, 0.32, 0.06 and 0.67 respectively. While for CH_4 it is found be 0.28, 0.71, 0.21, and 0.60 respectively. Negative correlation indicates that the influence of local sources on GHG~~'~~s, however, poor correlation coefficients during different seasons suggest the role of regional/local transport.
- CO_2 showed a positive correlation with temperature during all seasons except during winter. ~~Where as~~Whereas CH_4 showed a weak positive correlation with temperature during pre-monsoon and post-monsoon, while showing a weak negative correlation during monsoon and winter.
- CO_2 and CH_4 showed a strong positive correlation during winter, pre-monsoon, monsoon and post-monsoon with Rs equal to 0.80, 0.80, 0.61 and 0.72 respectively. This clearly

Formatted: Font: (Default) Times New Roman, 12 pt indicates common anthropogenic sources for these gases.~~the seasonal variations in source-sink mechanisms of CO_2 and CH_4 respectively.~~

- ~~Presence of OH radicals has been implicitly confirmed as a major sink of CH_4 over the study region.~~

Acknowledgment

This work was part of the Atmospheric CO_2 Retrieval and Monitoring (ACRM) under National Carbon Project (NCP) of ISRO-GBP. Authors sincerely acknowledge Mr. Biswadip Gharai, ACSG/ECSA for providing LULC data and to Mr. Mallikarjun, ACSG/ECSA for his support in data collection. We thank D & PQE division of NRSC and Mrs. Sujatha P, ACSG for sharing AWS and boundary layer data. The authors are grateful to the AT-CTM project of ISRO-GBP for providing the O_3 and NO_x analyzers. We would also like to thank HYSPLIT, ECMWF-ERA, MODIS and COSMIC team for providing scientific data sets used in this study. We also thankful to anonymous referees and the editor for providing constructive suggestions which certainly improved the quality of manuscript.

References

Ao, C. O., Waliser, D. E., Chan, S. K., Li, J. L., Tian, B., Xie, F., & Mannucci, A. J. Planetary boundary layer heights from GPS radio occultation refractivity and humidity profiles. Journal of Geophysical Research: Atmospheres (1984–2012), 117(D16), (2012).

Aurela M, Lohila A, Tuovinen JP, Hatakka J, Riutta T, Laurila T (2009) Carbon dioxide exchange on a northern boreal fen. Boreal Environment Research 14(4): 699-710

Baer, D. S., Paul, J. B., Gupta, M., & O'Keefe, A. Sensitive absorption measurements in the near-infrared region using off-axis integrated cavity output spectroscopy. In International Symposium on Optical Science and Technology (pp. 167-176). International Society for Optics and Photonics, (2002).

Berman, E. S., Fladeland, M., Liem, J., Kolyer, R., & Gupta, M. Greenhouse gas analyzer for measurements of carbon dioxide, methane, and water vapor aboard an unmanned aerial vehicle. Sensors and Actuators B: Chemical, 169, 128-135, (2012).

Chen, H., Wu, N., Wang, Y, & Peng, C. Methane is an Important Greenhouse Gas. Methane Emissions from Unique Wetlands in China: Case Studies, Meta Analyses and Modelling, chapter 1, (2015).

Crutzen, P. J., & Andreae, M. O. Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles. Science, 250(4988), 1669-1678, (1990).

Crutzen, P. J., A. C. Delany, J. Greenberg, P. Haagenson, L. Heidt, R. Lueb, W. Pollock, Wartburg Seiler, A. Wartburg, and P. Zimmerman. "Tropospheric chemical composition measurements in Brazil during the dry season." Journal of Atmospheric Chemistry 2, no. 3 (1985): 233-256.
Draxler RR, Rolph GD. HySPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY website (http://www.arl.noaa.gov/ready/hysplit4.html), NOAA Air Resources Laboratory. *Silver Spring, MD*. 2003.

Eisele, F. L., Mount, G. H., Tanner, D., Jefferson, A., Shetter, R., Harder, J. W., & Williams, E. J. Understanding the production and interconversion of the hydroxyl radical during the Tropospheric OH Photochemistry Experiment. Journal of Geophysical Research: Atmospheres (1984–2012), 102(D5), 6457-6465, (1997).

Fang, S. X., L. X. Zhou, P. P. Tans, P. Ciais, M. Steinbacher, L. Xu, and T. Luan. "In situ measurement of atmospheric CO_2 at the four WMO/GAW stations in China." Atmospheric Chemistry and Physics 14, no. 5 (2014): 2541-2554.

Fang, S. X., P. P. Tans, M. Steinbacher, L. X. Zhou, and T. Luan. "Study of the regional CO_2 mole fractions filtering approach at a WMO/GAW regional station in China." Atmospheric Measurement Techniques Discussions 8, no. 7 (2015).

~~Fearnside, Philip M. "Global warming and tropical land-use change: greenhouse gas emissions from biomass burning, decomposition and soils in forest conversion, shifting cultivation and secondary vegetation." Climatic change 46, no. 1-2 (2000): 115-158.~~

Frankenberg, Christian, Peter Bergamaschi, André Butz, Sander Houweling, Jan Fokke Meirink, Justus Notholt, Anna Katinka Petersen, Hans Schrijver, Thorsten Warneke, and Ilse Aben. "Tropical methane emissions: A revised view from SCIAMACHY onboard ENVISAT." Geophysical Research Letters 35, no. 15 (2008).

Garg, A., Bhattacharya, S., Shukla, P. R., & Dadhwal, V. K. Regional and sectoral assessment of greenhouse gas emissions in India. Atmospheric Environment, 35(15), 2679-2695, (2001).

Gaur, A., Tripathi, S. N., Kanawade, V. P., Tare, V., & Shukla, S. P. Four-year measurements of trace gases (SO_2, NOx, CO, and O_3) at an urban location, Kanpur, in Northern India. Journal of Atmospheric Chemistry, 71(4), 283-301, (2014).

Gilmanov, T. G., Johnson, D. A., Saliendra, N. Z., Akshalov, K., & Wylie, B. K. Gross primary productivity of the true steppe in Central Asia in relation to NDVI: scaling up CO2 fluxes. Environmental Management, 33(1), S492-S508, (2004).

Goroshi, S. K., Singh, R. P., Panigrahy, S., & Parihar, J. S. Analysis of seasonal variability of vegetation and methane concentration over India using SPOT-VEGETATION and ENVISAT-SCIAMACHY data. Journal of the Indian Society of Remote Sensing, 39(3), 315-321, (2011).

Hassan, A. G. A. Diurnal and Monthly Variations in Atmospheric CO_2 Level in Qena, Upper Egypt. Resources and Environment, 5(2), 59-65, (2015).

Hayashida, S., Ono, A., Yoshizaki, S., Frankenberg, C., Takeuchi, W., & Yan, X. (2013). Methane concentrations over Monsoon Asia as observed by SCIAMACHY: Signals of methane emission from rice cultivation. *Remote Sensing of Environment*, *139*, 246-256.

Huang, J.*, W. Zhang, J. Zuo, J. Bi, J. Shi, X. Wang, Z. Chang, Z. Huang, S. Yang, B. Zhang, G. Wang, G. Feng, J. Yuan, L. Zhang, H. Zuo, S. Wang, C. Fu and J. Chou, 2008: An overview of the Semi-Arid Climate and Environment Research Observatory over the Loess Plateau, Advances in Atmospheric Sciences, 25(6), 1-16.

Huang, J., Yu, H., Guan, X., Wang, G., & Guo, R. (2015). Accelerated dryland expansion under climate change. Nature Climate Change, doi:10.1038/nclimate2837.

Ichoku, C., & Ellison, L. Global top-down smoke-aerosol emissions estimation using satellite fire radiative power measurements. Atmospheric Chemistry and Physics, 14(13), 6643-6667, (2014).

Intergovernmental Panel on Climate Change (IPCC). Climate Change: The IPCC Scientific Assessment, edited by J. T. Houghton, G. J. Jerkins and J. J. Ephraums. Cambridge University Press. New York, (IPCC, 1990).

James, M. E., & Kalluri, S. N. The Pathfinder AVHRR land data set: an improved coarse resolution data set for terrestrial monitoring. International Journal of Remote Sensing, 15(17), 3347-3363, (1994).

Jing, X., Huang, J., Wang, G., Higuchi, K., Bi, J., Sun, Y., ... & Wang, T. (2010). The effects of clouds and aerosols on net ecosystem CO 2 exchange over semi-arid Loess Plateau of Northwest China. *Atmospheric Chemistry and Physics*, *10*(17), 8205-8218.

Jones, C., McConnell, C., Coleman, K., Cox, P., Falloon, P., Jenkinson, D., & Powlson, D. Global climate change and soil carbon stocks; predictions from two contrasting models for the turnover of organic carbon in soil. Global Change Biology, 11(1), 154-166, (2005).

Keppler, F., Hamilton, J. T., Braß, M., & Röckmann, T. Methane emissions from terrestrial plants under aerobic conditions. Nature, 439(7073), 187-191, (2006).

Kim, H. S., Chung, Y. S., Tans, P. P., & Dlugokencky, E. J. Decadal trends of atmospheric methane in East Asia from 1991 to 2013. Air Quality, Atmosphere & Health, 8(3), 293-298, (2015).

King, M. D., Kaufman, Y. J., Menzel, W. P., & Tanre, D. Remote sensing of cloud, aerosol, and water vapor properties from the Moderate Resolution Imaging Spectrometer (MODIS). Geoscience and Remote Sensing, IEEE Transactions on, 30(1), 2-27, (1992).

Kirschke, Stefanie., Bousquet, Philippe., Ciais, Philippe., Saunois, Marielle., Canadell, Josep G., Dlugokencky, Edward J., Bergamaschi, Peter., Bergmann, Daniel., Blake, Donald R., Bruhwiler, Lori., Cameron-Smith, Philip., Castaldi, Simona., Chevallier, Frédéric., Feng, Liang., Fraser, Annemarie., Heimann, Martin., Hodson, Elke L., Houweling, Sander., Josse, Béatrice., Fraser, Paul J., Krummel, Paul B., Lamarque, Jean-François., Langenfelds, Ray L., Quéré, Corinne Le., Naik, Vaishali., O'Doherty, Simon.,Palmer, Paul I.,Pison, Isabelle.,Plummer, David., Poulter, Benjamin.,Prinn, Ronald G.,Rigby, Matt.,Ringeval, Bruno., Santini,Monia.,Schmidt, Martina.,Shindell, Drew T.,Simpson, Isobel J.,Spahni, Renato.,Steele, L. Paul.,Strode, Sarah A.,Sudo, Kengo.,Szopa, Sophie.,Werf, Guido R. van der.,Voulgarakis, Apostolos., Weele, Michielvan.,Weiss, Ray F.,Williams, Jason E, Guang, Zeng. Three decades of global methane sources and sinks, Nature Geoscience, volume 6, (2013), doi: 10.1038/NGEO1955.

Koshal. A. K. Spatial temporal climatic change variability of cropping system in western Uttar Pradesh, International Journal of Remote Sensing & Geoscience, volume 2, issue 3, (2013).

Lewis, A. C., Evans, M. J., Hopkins, J. R., Punjabi, S., Read, K.A., Purvis, R. M., Andrews, S. J., Moller, S. J., Carpenter, L.J., Lee, J. D., Rickard, A. R., Palmer, P. I., and Parrington, M.: The influence of biomass burning on the global distribution of selected non-methane organic compounds, Atmos. Chem. Phys., 13, 851–867, doi:10.5194/acp-13-851-2013, 2013.

Liu, Yang, Xiufeng Wang, Meng Guo, Hiroshi Tani, Nobuhiro Matsuoka, and Shinji Matsumura. "Spatial and temporal relationships among NDVI, climate factors, and land cover changes in Northeast Asia from 1982 to 2009." GIScience & Remote Sensing 48, no. 3 (2011): 371-393.

Machida, T., K. Kita, Y. Kondo, D. Blake, S. Kawakami, G. Inoue, and T. Ogawa. "Vertical and meridional distributions of the atmospheric CO2 mixing ratio between northern midlatitudes and southern subtropics." Journal of Geophysical Research: Atmospheres (1984–2012) 107, no. D3 (2002): BIB-5.

Mahesh, P., N. Sharma, V. K. Dadhwal, P. V. N. Rao, and B. V. Apparao. "Impact of Land-Sea Breeze and Rainfall on CO2 Variations at a Coastal Station. J Earth Sci Clim Change 5: 201. doi: 10.4172/2157-7617.1000201 Volume 5. Issue 6. (2014).

Mahesh. P, Sreenivas. G, Rao.P.V.N., Dadhwal.V.K.,Sai Krishna. S.V.S. and Mallikarjun. K: High precision surface level CO_2 and CH_4 using Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS) over Shadnagar, India, International Journal of Remote Sensing, (2015), doi:10.1080/01431161.2015.1104744.

Miller, John B., Luciana V. Gatti, Monica TS d'Amelio, Andrew M. Crotwell, Edward J. Dlugokencky, Peter Bakwin, Paulo Artaxo, and Pieter P. Tans. "Airborne measurements indicate large methane emissions from the eastern Amazon basin." Geophysical Research Letters 34, no. 10 (2007).

Monastersky, Richard. "Global carbon dioxide levels near worrisome milestone." Nature 497, no. 7447 (2013): 13-14.

Nair, V. S., Moorthy, K. K., Alappattu, D. P., Kunhikrishnan, P. K., George, S., Nair, P. R., & Niranjan, K. (2007). Wintertime aerosol characteristics over the Indo-Gangetic Plain (IGP): Impacts of local boundary layer processes and long-range transport. Journal of Geophysical Research: Atmospheres (1984–2012), 112(D13).

Newman, S., Jeong, S., Fischer, M. L., Xu, X., Haman, C. L., Lefer, B., Alvarez, S.,Rappenglueck, B.,Kort, E. A., Andrews, A. E., Peischl, J.,Gurney, K. R.,Miller, C. E.,and Yung, Y. L.,: Diurnal tracking of anthropogenic CO_2 emissions in the Los Angeles basin megacity during spring, Atmos. Chem. Phys., 13, 4359–4372, 2013, doi:10.5194/acp-13-4359-2013.

Nishanth, T., K. M. Praseed, M. K. Satheesh Kumar, and K. T. Valsaraj. "Observational study of surface O3, NOx, CH4 and total NMHCs at Kannur, India." Aerosol. Air. Qual. Res 14 (2014): 1074-1088.

Pan, X. L., Kanaya, Y., Wang, Z. F., Liu, Y., Pochanart, P., Akimoto, H., Sun, Y. L., Dong, H. B., Li, J., Irie, H., and Takigawa, M.: Correlation of black carbon aerosol and carbon monoxide in the high-altitude environment of Mt. Huang in Eastern China, Atmos. Chem. Phys., 11, 9735-9747, doi:10.5194/acp-11-9735-2011, 2011

Paul, J. B., Lapson, L., & Anderson, J. G.. Ultrasensitive absorption spectroscopy with a high-finesse optical cavity and off-axis alignment. Applied Optics, 40(27), 4904-4910, (2001).

Patil, M. N., T. Dharmaraj, R. T. Waghmare, T. V. Prabha, and J. R. Kulkarni. "Measurements of carbon dioxide and heat fluxes during monsoon-2011 season over rural site of India by eddy covariance technique." Journal of Earth System Science 123, no. 1 (2014): 177-185.

Pielke, Roger A., Gregg Marland, Richard A. Betts, Thomas N. Chase, Joseph L. Eastman, John O. Niles, and Steven W. Running. "The influence of land-use change and landscape dynamics on the climate system: relevance to climate-change policy beyond the radiative effect of greenhouse gases." Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences 360, no. 1797 (2002): 1705-1719.

Ramachandran, S., and T. A. Rajesh. "Black carbon aerosol mass concentrations over Ahmedabad, an urban location in western India: comparison with urban sites in Asia, Europe, Canada, and the United States." Journal of Geophysical Research: Atmospheres (1984–2012) 112, no. D6 (2007).
Salomonson, Vincent V., W. L. Barnes, Peter W. Maymon, Harry E. Montgomery, and Harvey Ostrow. "MODIS: Advanced facility instrument for studies of the Earth as a system." Geoscience and Remote Sensing, IEEE Transactions on 27, no. 2 (1989): 145-153.

Smith, K. A., Ball, T., Conen, F., Dobbie, K. E., Massheder, J., & Rey, A. Exchange of greenhouse gases between soil and atmosphere: interactions of soil physical factors and biological processes. European Journal of Soil Science, 54(4), 779-791, (2003).

Schneising, O., M. Buchwitz, J. P. Burrows, H. Bovensmann, P. Bergamaschi, and W. Peters. "Three years of greenhouse gas column-averaged dry air mole fractions retrieved from satellite– Part 2: Methane." Atmos. Chem. Phys 9, no. 2 (2009): 443-465.

Sharma Neerja, Dadhwal, V.K., Kant, Y., Mahesh, P., Mallikarjun, K., Gadavi, Harish, Sharma, Anand., Ali, M.M. Atmospheric CO_2 Variations in Two Contrasting Environmental Sites Over India. Air, Soil and Water Research 2014:7 61–68, (2014), doi:10.4137/ASWR.S13987.

Sharma, Anu Rani, Shailesh Kumar Kharol, K. V. S. Badarinath, and Darshan Singh. "Impact of agriculture crop residue burning on atmospheric aerosol loading—a study over Punjab State, India." In Annales geophysicae: atmospheres, hydrospheres and space sciences, vol. 28, no. 2, p. 367. 2010.

Sharma, Neerja, Rabindra K. Nayak, Vinay K. Dadhwal, Yogesh Kant, and Meer M. Ali. "Temporal variations of atmospheric CO_2 in Dehradun, India during 2009." Air, Soil and Water Research 6 (2013): 37.

Stocker, T.F., Qin, D., Plattner, G.K., Alexander, L.V., Allen, S.K., Bindoff, N.L., Bréon, F.M., Church, J.A., Cubasch, U., Emori, S., Forster, P., Friedlingstein, P., Gillett, N., Gregory, J.M., Hartmann, D.L., Jansen, E., Kirtman, B., Knutti, R., Krishna Kumar, K., Lemke, P., Marotzke, J., Masson-Delmotte, V., Meehl, G.A., Mokhov, I.I., Piao, S., Ramaswamy, V., Randall, D., Rhein, M., Rojas, M., Sabine, C., Shindell, D., Talley, L.D., Vaughan D.G., and Xie, S.P. Technical Summary. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, (2013).

Shea, S.J.O, G. Allen1, M. W. Gallagher, S. J.-B. Bauguitte, S. M. Illingworth, M. Le Breton, J. B. A. Muller,C. J. Percival, A. T. Archibald, D. E. Oram, M. Parrington,*, P. I. Palmer, and A. C. Lewis. Airborne observations of trace gases over boreal Canada during BORTAS: campaign climatology, air mass analysis and enhancement ratios. Atmos. Chem. Phys., 13, 12451–12467, 2013 atmos-chem-phys.net/13/12451/2013/ doi:10.5194/acp-13-12451-2013

Stohl, Andreas, Markus Hittenberger, and Gerhard Wotawa. "Validation of the Lagrangian particle dispersion model FLEXPART against large-scale tracer experiment data." Atmospheric Environment 32, no. 24 (1998): 4245-4264.

Stull, R. B. (1988). Similarity theory. In *An Introduction to Boundary Layer Meteorology* (pp. 347-404). Springer Netherlands.

Taylor J. An Introduction to Error Analysis: The Study of Uncertainties in Physical Measurement, University Science Books, And ISBN: 093570275X (ISBN13: 9780935702750), (1997).

Thum, T., T. Aalto, T. Laurila, M. Aurela, J. Hatakka, Anders Lindroth, and T. Vesala. "Spring initiation and autumn cessation of boreal coniferous forest CO2 exchange assessed by meteorological and biological variables." Tellus B 61, no. 5 (2009): 701-717.

Tohjima, Y., Kubo, M., Minejima, C., Mukai, H., Tanimoto, H., Ganshin, A., Maksyutov, S., Katsumata, K., Machida, T., and Kita, K.: Temporal changes in the emissions of CH4 and CO from China estimated from CH4 / CO2 and CO / CO2 correlations observed at Hateruma Island, Atmos. Chem. Phys., 14, 1663-1677, doi:10.5194/acp-14-1663-2014, 2014.

Vaghjiani, Ghanshyam L., and A. R. Ravishankara. "New measurement of the rate coefficient for the reaction of OH with methane." Nature 350, no. 6317 (1991): 406-409.

Wang, B-R., X-Y. Liu, and J-K. Wang. "Assessment of COSMIC radio occultation retrieval product using global radiosonde data." Atmospheric Measurement Techniques 6, no. 4 (2013): 1073-1083.

Wang, G., J. Huang*, W. Guo, J. Zuo, J. Wang, J. Bi, Z. Huang, and J. Shi, 2010: Observation analysis of land-atmosphere interactions over the Loess Plateau of northwest China, J. Geophys. Res., 115, D00K17,doi:10.1029/2009JD013372.

Worthy, Douglas EJ, Elton Chan, Misa Ishizawa, Douglas Chan, Christian Poss, Edward J. Dlugokencky, Shamil Maksyutov, and Ingeborg Levin. "Decreasing anthropogenic methane emissions in Europe and Siberia inferred from continuous carbon dioxide and methane observations at Alert, Canada." Journal of Geophysical Research: Atmospheres (1984–2012) 114, no. D10 (2009).

Yunck, Thomas P., Liu Chao-Han, and Randolph Ware. "A history of GPS sounding." Terrestrial Atmospheric and Oceanic Sciences 11, no. 1 (2000): 1-20.

Table 1 Data used

Sensor	Period	Parameter	resolution	Source
GGA-24EP	Jan-2014 to Dec 2014	CO_2,CH_4 and H_2O	1 Hz time	ASL,NRSC
42*i*-NO-NO_2-NO_x	Jul-2014 to Sep-2014	NOx(=NO+NO_2)	1 min time	ASL,NRSC
49i-O_3	Jul-2014 to Sep-2014	O_3	1 min time	ASL,NRSC
AWS	Jan-2014 to Dec-2014	WS,WD,AT,RH	60 min time	NRSC
Terra/MODIS	Jan-2014 to Dec-2014	NDVI	5 Km horizontal	http://ladsweb.nascom.nasa.gov/data/search.html
COSMIC-1DVAR	Jul-2013 to Jun-2014	Refractivity (N)	0.1 Km vertical	
HYSPLIT	Jan-2014 to Dec-2014	Backward trajectory	5 day isentropic model (1km to 4 km)	http://ww.arl.noaa.gov/ready/hysplit4.html]
FEER v1	Jan-2013 to Dec-2013	fire radiative power (FRP)		http://ladsweb.nascom.nasa.gov/data/search.html

Table 2 Statistical correlation between CO_2 and CH_4

S.No	**Seasons**	**Correlation coefficient (R)**	**Slope** $\left(\frac{Y_{CH4}\,(ppm)}{X_{CO2}(ppm)}\right)$	Ψ_{slope} **(ppm)**	$\Psi_{y\text{-}int}$ **(ppm)**
1	Monsoon (JJAS)	0.61	0.005	0.00015	1.91
2	Post-monsoon (OND)	0.72	0.0065	0.00014	1.52
3	Winter (JF)	0.80	0.0085	0.00018	9.13
4	Pre-monsoon (MAM)	0.80	0.0059	0.00021	2.73

Table ~~2~~3 Seasonal amplitudes of CO_2 and CH_4 over study region arriving from different directions

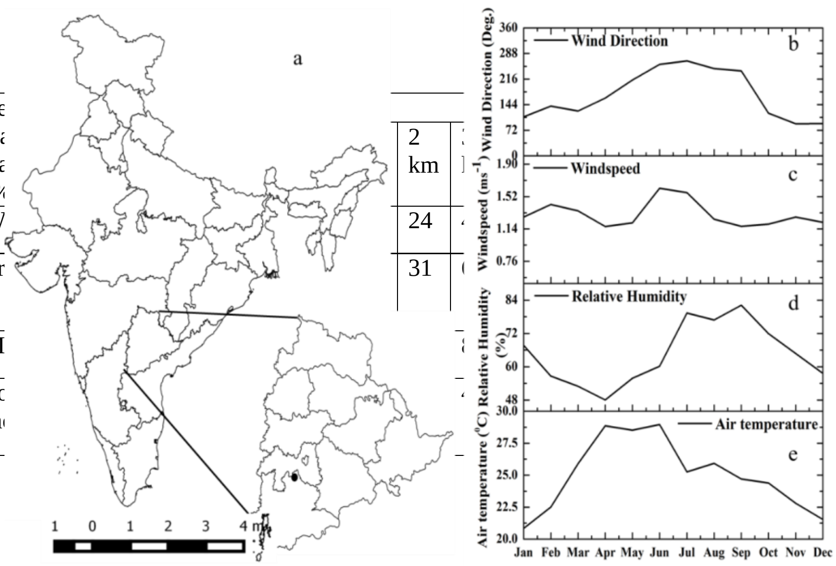
Wind Direction	Winter $\frac{CO_2}{CH_4}$ (ppm)	Pre-monsoon $\frac{CO_2}{CH_4}$ (ppm)	Monsoon $\frac{CO_2}{CH_4}$ (ppm)	Post-monsoon $\frac{CO_2}{CH_4}$ (ppm)
0-45	399.85/1.98	410.37/1.94	400.72/1.91	395.13/2.02
45-90	391.66/1.94	399.59/1.89	388.82/1.91	390.23/1.98
90-135	391.57/1.93	397.79/1.87	388.99/1.87	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

~~**Table 3** Statistical correlation between~~ ~~CO_2~~ ~~and~~ ~~CH_4~~

S.No	Seasons	Correlation coefficient (R^2)	Slope ($\frac{Y_{CH4}(ppm)}{X_{CO2}(ppm)}$)	ψ_{slope} (ppm)	$\Psi_{y\text{-}int}$ (ppm)
1	Monsoon (JJAS)	0.6137	0.005	0.00015	1.91
2	Post-monsoon (OND)	0.7252	0.0065	0.00014	1.52
3	Winter (JF)	0.8061	0.0085	0.00018	9.13
4	Pre-monsoon (MAM)	0.8064	0.0059	0.00021	2.73

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

Se Ba tra (%	2 km	[illegible]	2 km	3 km	4 km
W	24	[illegible]	19	42	41
Pr	31	[illegible]	14	26	11
M	[illegible]	[illegible]	61	9	5
Pc m	[illegible]	[illegible]	2	16	11



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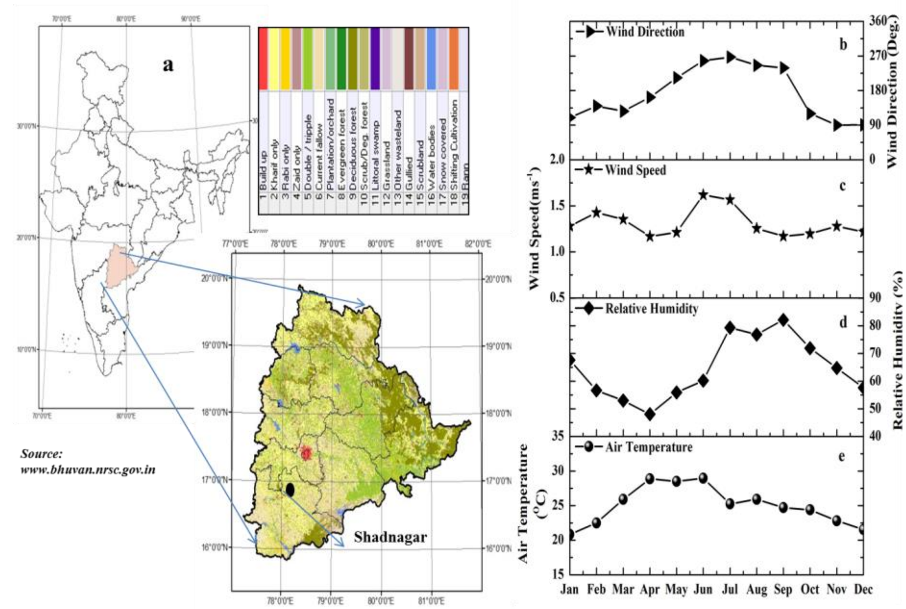

Figure 1 a) Schematic representation of study area; b-e) Seasonal variation of prevailing meteorological conditions during 2014~~study period~~

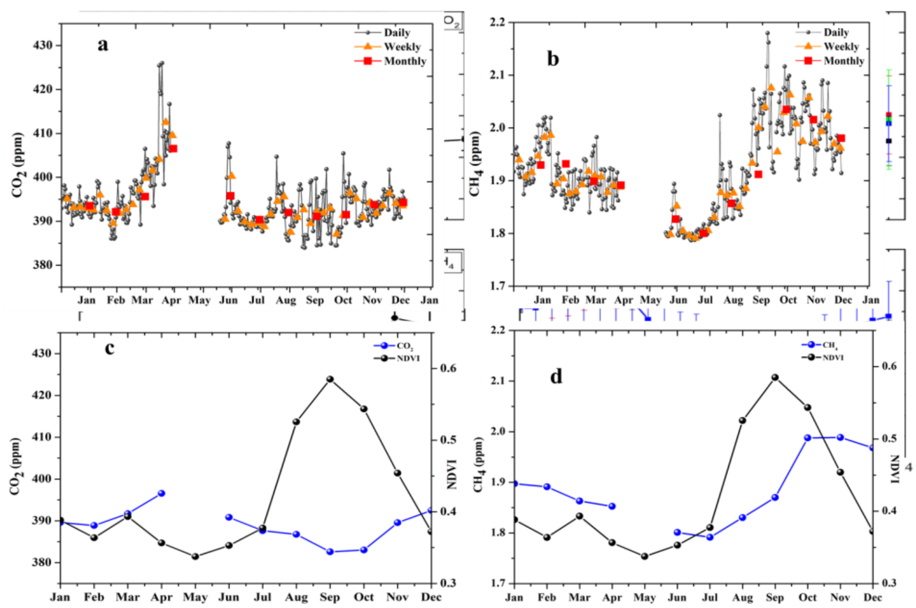

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Figure 2 a-b) Temporal Seasonal variations of CO_2 and CH_4; c-d) Seasonal variations of CO_2 and CH_4 in conjunction with NDVI (Normalized Difference Vegetation Index)diurnal variations of CO_2 and CH_4 during 2014

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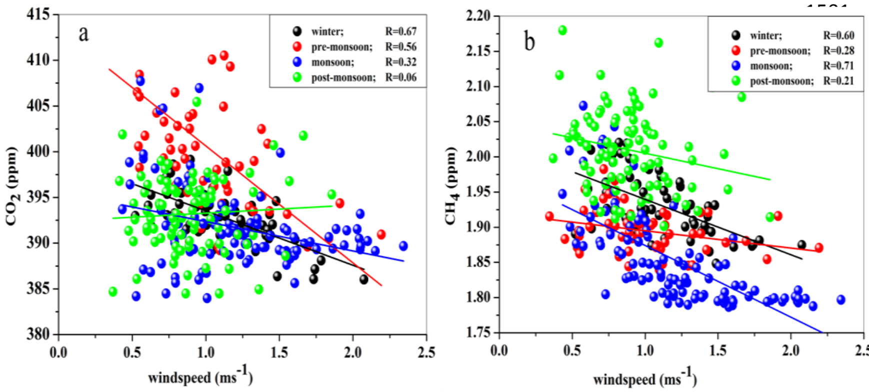

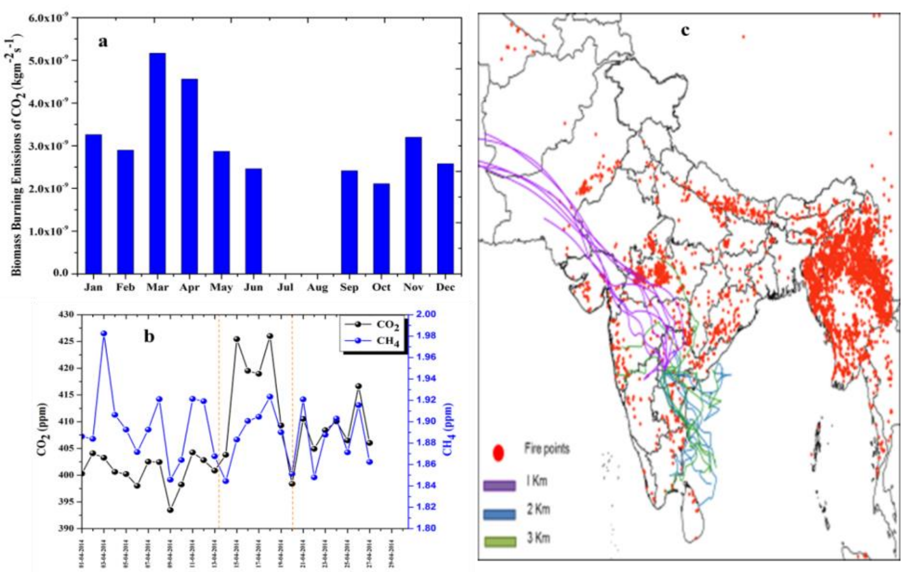

Figure 3 ~~a-b)~~ a) Long term analysis of CO_2 biomass burning emissions over study region b) Biomass signatures on CO_2/CH_4 during 14-21 April 2014, a case study c) Spatial distribution of MODIS derived fire counts over Indian region during 14-21 April 2014.~~Scatterplot between wind speed and GHG's (CO_2 and CH_4).~~

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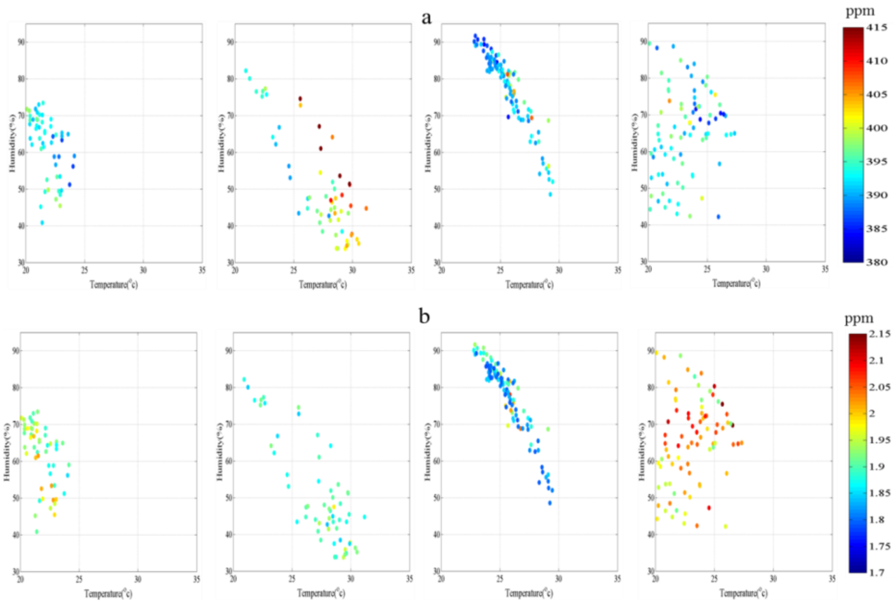

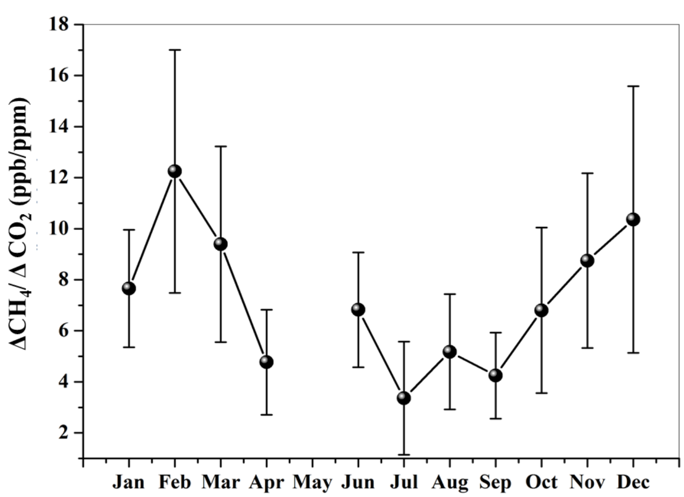

Figure 4 Monthly variation of $\Delta CH_4/\Delta CO_2$ during study period

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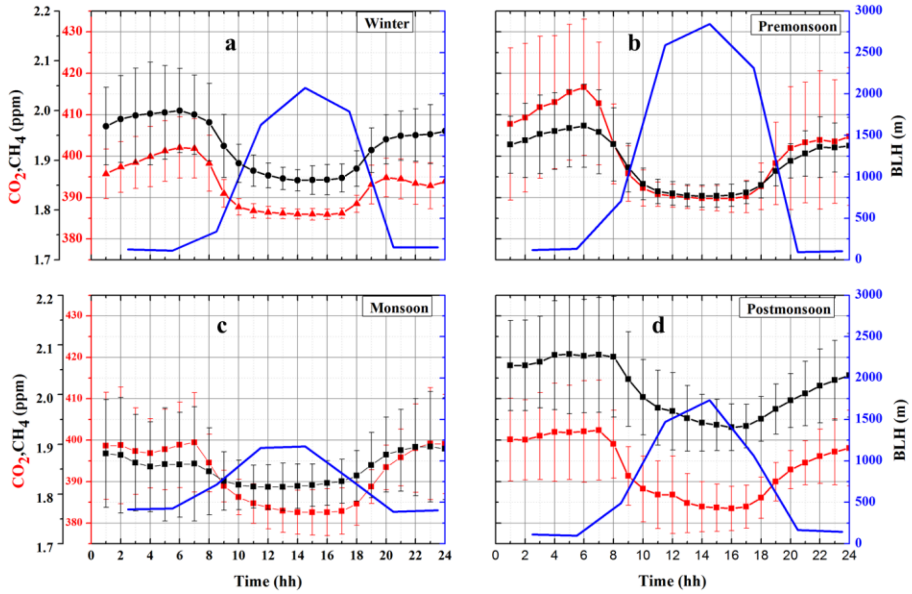

Figure 5 a-d) Seasonal variations of diurnal averaged CO_2/CH_4 against boundary layer height during 2014

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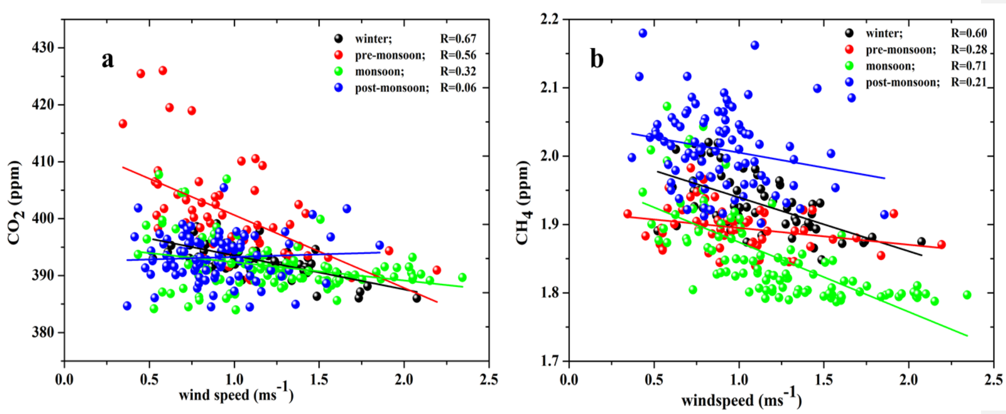

Figure 6 a-b) Daily mean scatterplot between wind speed and GHGs (CO_2 and CH_4).

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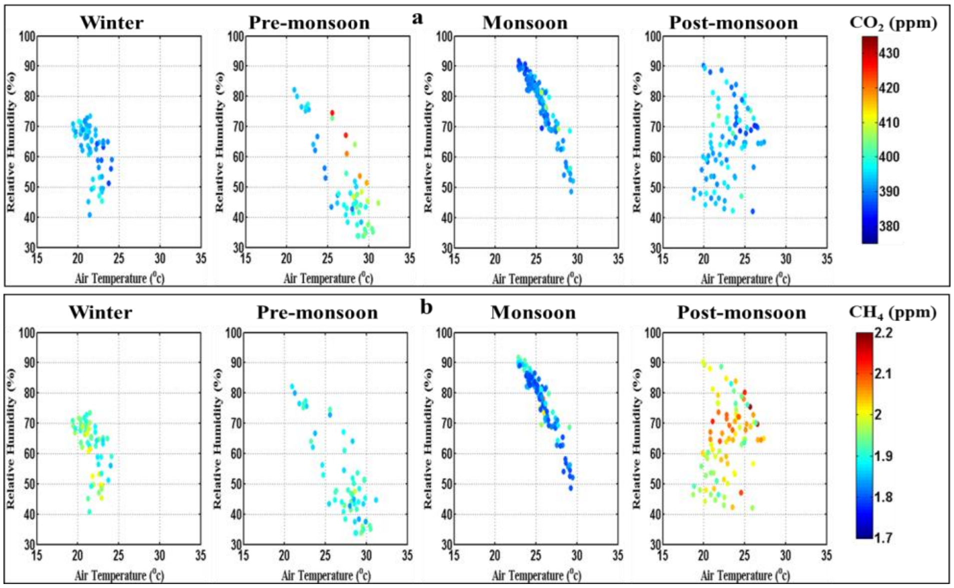

Figure 7 a-b) Daily mean seasonal variation of CO_2 and CH_4 as function of humidity and air temperature during 2014

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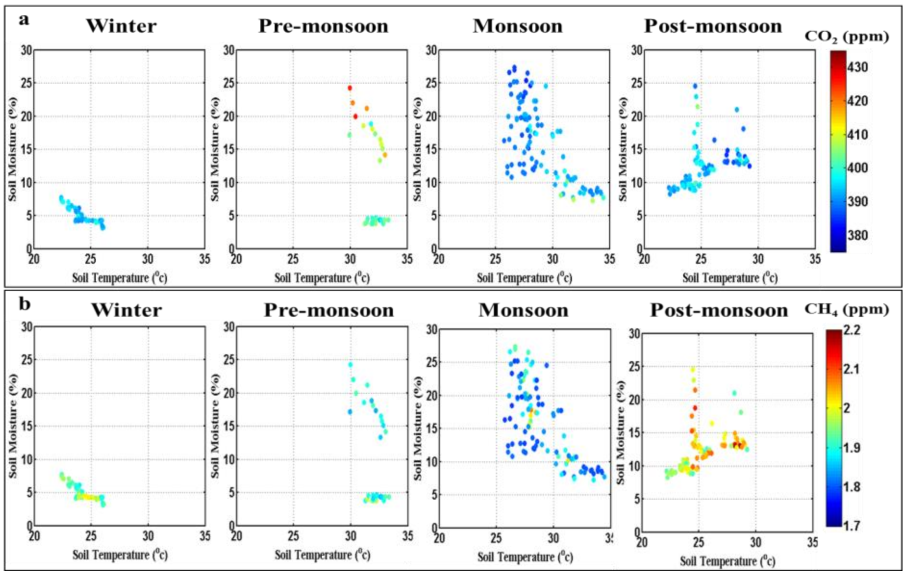

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Figure 8 a-b) Daily mean seasonal variation of CO_2 and CH_4 as function of soil temperature and soil moisture during 2014a) Seasonal variation of CO_2 as function of humidity and temperature during winter, pre-monsoon, monsoon and post-monsoon. b) Seasonal variation of CH_4 as function of humidity and temperature during respective seasons

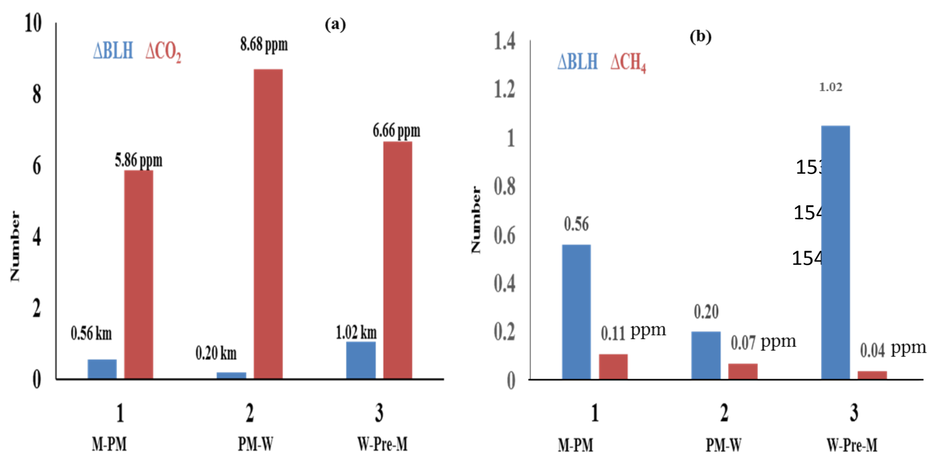


Figure 59 Seasonal difference in BLH variations ofagainst respective change in -a) CO_2 and b) CH_4 against boundary layer height change

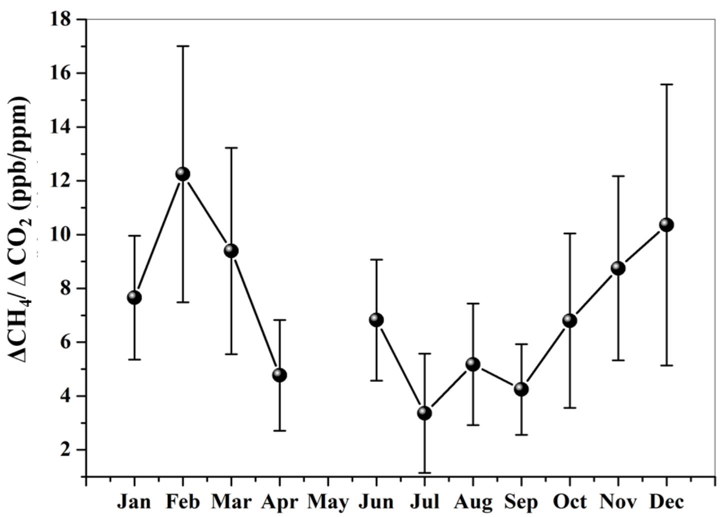


Figure 6 Monthly variation of $\Delta CH_4/\Delta CO_2$ during study period

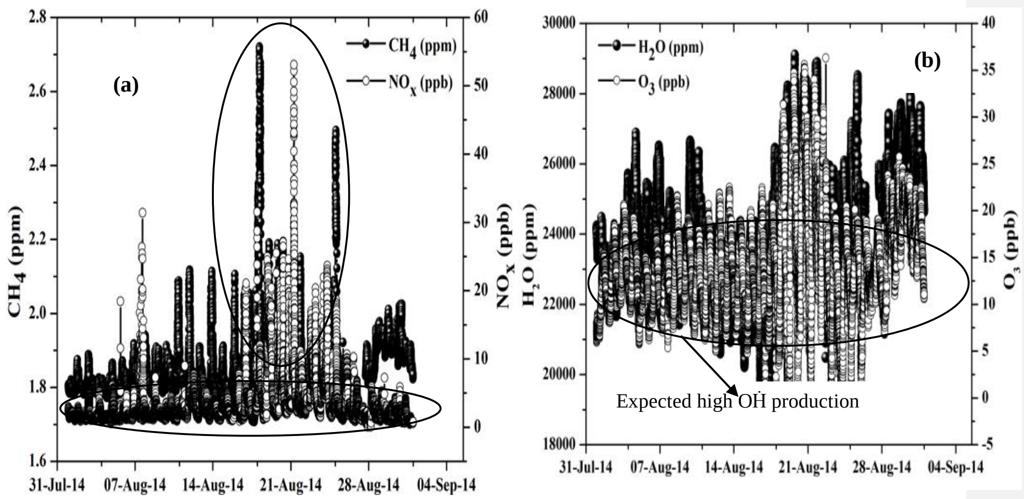
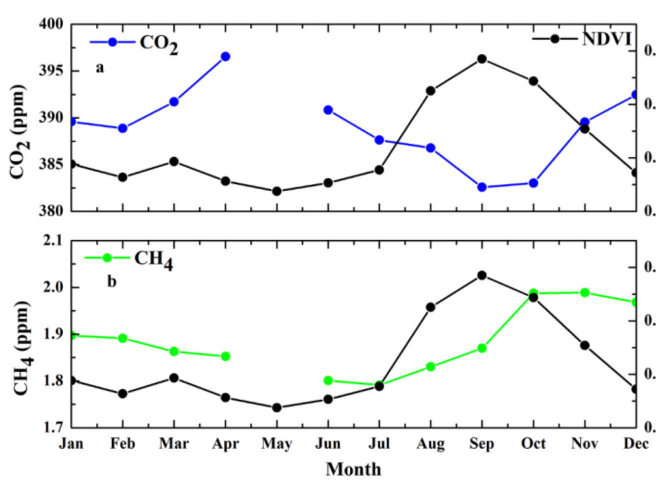


Figure ~~7~~10 Time series analysis of a) CH_4 vs. NO_x, b) H_2O vs. O_3



~~**Figure 8** a) Seasonal variation of CO_2 in conjunction with NDVI (Normalized Difference Vegetation Index). b) Seasonal variation of CH_4 in conjunction with NDVI~~

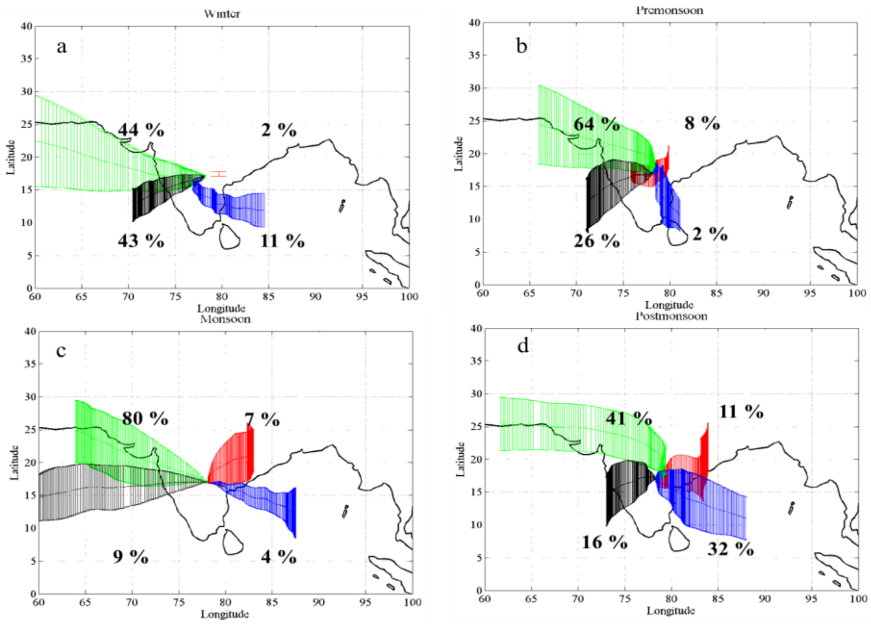


Figure ~~9~~11 a-d) Long range circulation of air mass trajectories ending over Shadnagar at 3 km during winter, pre-monsoon, monsoon and post-monsoon

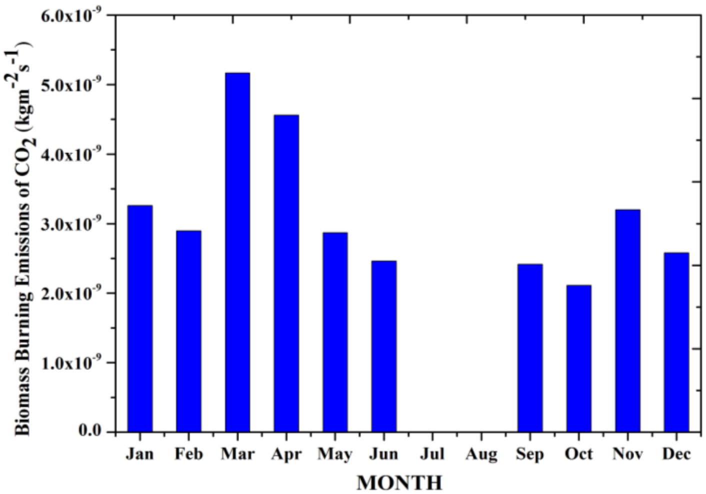


Figure 10 Long term analysis of CO_2 biomass burning emissions over study region