Measurement report: Regional characteristics of seasonal and longterm variations in greenhouse gases at Nainital, India and Comilla, Bangladesh, by S. Nomura, M. Naja, M. K. Ahmed, H. Mukai, Y. Terao, T. Machida, M. Sasakawa, and P. K. Patra

Response to Reviewers

We would like to thank the reviewers for providing comments and suggestions in our manuscript. We revised the manuscript based on the comments. Comments and questions from reviewers are reproduced here in black. Responses to reviewers are written in red.

Anonymous Referee #4

This study presents GHG observations over Northern Indian sites of Nainital NTL and Comilla CLA, Bangladesh. Factors like transported airmass, local cropping, biomass burning and precipitation locally seem to play a role in the observed variability at these sites. CLA show overall high CH4 concentration throughout the year. On the other hand, SF6 concentrations are similar to that at MLO, suggests that not many urban activities or anthropogenic emissions are active near these sites. This study emphasis that Indian Dipole DMI affects circulation and precipitation which in turn affects the growth rates of GHGs.
NTL and CLA long-term observations can play an important role towards understanding the regional carbon budget over South Asia. GHG variability in terms of seasonality, airmass transport dynamics, are already reported in various studies in the past (papers are cited in this study). However, studies reporting carbon flux estimation using top-down modelling are limited over this region.
Observations presented in this study are very useful to understand carbon budget over South Asia. NTL and CLA observed data should be available on public domain for other researchers at the earliest. This manuscript may be accepted for publication in ACP after replying following comments.

>Thank you very much for seeing the value of our observations. We appreciate your constructive comments and suggestions.

1) L21-21: NTL do not show minima in Feb-March (ref. Fig. 6)

>We removed “NTL” in L21 and added the sentence in L22-23 as follows: “Although NTL had only one clear minima in September,…”.

2) L25: “….in addition to other sources..” , what are other sources, pls specify.

>Biomass burning would be partly contributed to high CH_4 in August–October but we need further studies for estimates of contributions from biomass burnings. We removed "in addition to other sources" and modified the sentence as "mainly due to the influence of CH_4 emissions from the paddy fields." in L25-26.

3) L26-27: "High CH4 mole fractions……Plain", Is it due to large scale airmass transport or local emission?

>High CH_4 mole fractions were affected by the both local emission and air mass transport over Indo-Gangetic Plain. We added the sentence: "which were affected by the both local emission and air mass transport." in L27.

4) L32-33: SF6 mole fraction is similar to that at MLO this suggests that there are few anthropogenic emissions sources near those places. However, CO observations are high at both the sites. Is it not a that a contradictory result?

>SF_6 is used mainly for high voltage equipment such as step-up or down transformer and electrical plants. Some portion will be come from the process of its production. But CO is emitted mainly from the biomass burning. Thus, SF_6 emission source is completely different from the CO emission source. We modified the sentence of "there were few anthropogenic emission sources" to "there were few anthropogenic SF_6 emission sources" in L33.

5) L49-50: "…because there are few measured GHG mole fractions in the South Asian region" ; "Several observations on GHG mole fractions in the atmosphere have been done around India"…….Two contradictory statement. Consider revising.

>We changed the expression of "few" to "only a few". We removed the sentence of "Several observations on GHG mole fractions in the atmosphere have been done around India" in L50.

6) L56-59: Do you mean CH4 and CO sources are co-located over these regions. Consider revising text in these lines.

>This paper didn't indicate that CH4 and CO sources are co-located over these regions. This paper just indicates that Indian subcontinent has strong emission sources of CH4 and CO and the atmospheric mole fractions of CH4 and CO is affected by the seasonal wind. Major CH4 emission source and major CO emission source is different from the results of our flask sampling. Major CH4 emission source is

the paddies field and the major CO emission source is the biomass burning.

7) L83-84: “Thus the GHG observations ……..long-term trend remain limited”. This sentence is not clear. What do you men by long term trend remail limited? Consider revising.

>We modified the sentence: “Thus, the GHGs observation program in Indian region is expanding gradually, however, the characterization of GHGs behaviour in the northern Indian subcontinent and their long-term trends are not well understood.”. in L83.

8) L84-94: “In this work ……..ENSO index”. Why this study is important and how it fills gap areas left behind from past studies. Consider revising this paragraph.

>We added the sentence of “In this paper, we present the longer GHGs data than previous studies in the Indo-Gangetic Plain including Bangladesh, which is a blank area for GHGs observation and clarify the characteristics of GHGs in the Indian subcontinent by analyzing the periodicity of GHGs growth rates and comparing them with regional climatic conditions.” in L84-87.

9) L100: “…Mt. Mauna Peak….”, is it Manora Peak? Pls check and correct.

>We modified “Mt. Mauna Peak” to Mt. Manora Peak” in L103 as your suggestion.

10) L103-104: “We estimated that …….nearby”, have you estimated or assumed? If you estimated then what is the basis for estimation? Same for assumption

>We modified “which mean that NTL might be influenced mainly by the air mass passing through the Indo-Gangetic Plain. We estimated that the air of NTL is not strongly influenced by local GHGs emissions nearby.” to “which mean that the air of NTL is influenced mainly by the air mass passing through the Indo-Gangetic-Plain, rather than extremely influenced by local GHGs emissions nearby.” in L105-107.

11) L107-109: “Farmers in Comilla ……nearby emissions”, this indicates that CLA is strongly influenced by the local anthropogenic emissions. On the other hand, based on SF6 observations you say that these sites are free from local emissions (ref. abstract). Its better to be consistent with the site characteristics described in the text. Also, be consistent in mentioning site name. Use either Comilla or CLA.

>SF_6 is not emitted by the biomass burning by the farmer. We modified "Comilla" to "CLA" in L109.

12) L112: "…..(on the roof of the second floor of the station) in NTL…….", What is height of the inlet head from the roof surface? What is height of the canopy close to the inlet head?

>Height of the inlet is 1-m higher than the roof surface. We added the information of the height of the inlet of NTL to the canopy in L117-118.

13) L113: What sealing material used in Pyrex flasks? Is it comparable to the boro 3.3 flasks (from Normag) and PCTFE sealing material used at MPI Jena.

>We use the Viton O-rings for sealing. We added the information of sealing in L 122.

14) L121: Air samples were cooled at -30 dc while sampling at NTL and CLA. Again, they are cooled at -80 dc before injecting to the analytical system at NIES. An explanation should given about this. Whether cooling twice (double dehumidification) have any scientific basis?

>To dry air samples almost completely before analysis is essential, because CO_2 and other GHGs should be measured on dry air base. So, we use -80dc coolant basically for our analytical system. But for sampling, we just use rather simple cooling system to prevent water from condensing on the inner surface of the glass flask.

15) L122: Fig.2b should be simplified for the ease of readers. Put the direction of sample flow. Too many text inside the figure makes it complicated. Avoid writing text such as "Peak labs, Peak Performer, Agilent 7890, etc." inside this figure. It may be mentioned in the figure caption.

>We added the sample flow to the Fig.2b.
We removed "Peak labs, Peak Performer, Agilent 7890, etc" in the Fig.2b.

16) L126-127: "….GC-ECD or GC-micro-ECD", which one is used exactly?

>We modified the sentence "a gas chromatograph with an electron capture detector or a micro electron capture detector (GC-ECD or GC-micro-ECD" to "a gas chromatograph with an electron capture detector (GC-ECD) until 2011 and with a micro electron capture detector (GC-micro-ECD) from 2012" in L134-135.

17) L135-149: A figure may be shown similar like Fig.2b

>The analysis line of GHGs mole fraction in the NIES laboratory is complex and consists of multiple instruments. We added the schematic because it contributes to understand the analysis line of GHGs mole fraction in the NIES laboratory. While, the analysis line of isotopic ratio in the NIES laboratory is simple and consists of one instrument. So, we judged that the schematic of the analysis line of isotopic ratio in the NIES laboratory isn't necessary in this paper.

18) L151-158: MLO is a reference site, however CRI does not represent a global/continental signal. It's a sub-regional site. Airmasses arrive at CRI are different than that of NTL and CLA. Hanle (HLE: French controlled site in India) or Seychelles (SEY) better represents large air masses in this region and can be considered as reference site. HLE represents northern hemisphere and SEY southern hemisphere. I suggest replacing CRI with HLE and SEY.

>We selected MLO and CRI for comparing with NTL and CLA because the data of MLO has representative of middle latitude of northern hemisphere and the data of CRI includes the characteristic of GHGs of India subcontinent. The mole fraction of GHGs in CRI shows the same level with the data of MLO when the air mass transported from Indian Ocean, while the mole fraction of GHGs in CRI shows the high concentration when the air mass transported over the India subcontinent. We didn't select SEY and HLN for comparing with NTL and CLA because the data of SEY were similar to the data of MLO and the SEY was located in southern hemisphere. We didn't use HLE because it is located at north area of Himalaya Mountain: We need to select the comparison site in south area from the Himalaya Mountain because the north area and the south area of Himalaya Mountain are quite different about the transportation of air mass, terrestrial condition and anthropogenic activities.

19) L176-177: Give more details about calculating the ratio.

>We modified "the ratio of air mass from south was calculated by the frequency of the air mass from south side on the flask sampling date with reference to the backward air trajectories data." to "the ratio of air mass from south per year was calculated by the frequency of the air mass from south side of Indian Ocean on the flask sampling date in each year with reference to the backward air trajectories data calculated by METEX." in L192-193.

20) L180: How you supplemented the value of missing period and any error in it. Describe in detail.

>The value of the missing period was supplemented with an interpolated values from the previous and

following data of the missing period for calculating the continuous long-term trend and smoothing fitting curve. We described in the text.

21) L191: Consider revising title of section 3.1, use of word “levels” may be misleading. May be replaced with concentration and low-concentration, high-concentration in the text.

>We replaced “level” to “mole fraction” or its “values”.

22) Fig. 4: MLO curve clearly not visible after 2013 onward. Also, I do not understand the scientific reason behind using CRI data here. It is used because data is freely available at WDCGG? HLE would have been better background site like MLO. CRI is not advisable to use as a reference unless strong scientific motive is described.

>The mole fraction of GHGs in CRI shows the same level with the data of MLO when the air mass transported from Indian Ocean, while the mole fraction of GHGs in CRI shows the high concentration when the air mass transported over the India subcontinent. The data of CRI are very helpful in characterizing the behavior of GHGs mole fraction in India subcontinent. The data of HLE doesn’t clear the seasonal variation of GHGs by the monsoon in India subcontinent. Also, we got the data of CRI on the WDCGG web site with contact to CSIRO staff.

23) L223-224: “….CRI site represents Southern Hemisphere during JJAS…”, in that case you can use Seychelles (SEY) which is better representative of southern hemisphere.

>It is important that CRI is located in India subcontinent. We wanted to indicate that the data of CRI is influenced by the air mass from southern hemisphere during monsoon season only and same level of mole fraction with the data of MLO (also SEY) during monsoon season.

24) L227-233: CRI represents large part of Indian land mass during Nov-March. Oct and April are air mass transition months (seasonal change). So CRI site observations are good example of seasonal reversal of wind pattern. GHG mole fractions at CRI during JJAS represents oceanic air masses (pristine environment) and rest of the months it represents Indian land mass. Mole fractions representing Indian land mass may dominate in annual average. Such discussions should be written in the section 3.2.1.

>The same content is written in the first paragraph in the section.

25) L234-235: As mean growth rate (CO2) at NTL and CLA agrees with MLO, curves showing this should be added in Fig. 5a

>Thank you for your suggestion. We added the values of annual mean growth rate (CO2) at NTL and CLA in Table 1.

26) L238-252: NTL and CLA CO2 growth and its relationship with ENSO and IOD are discussed in these paragraphs. However, no such statement is made in the conclusions section. Please add few lines about this in conclusion section as well. I am very surprise to see that there is no relationship between NTL and ENSO index. As ENSO is global phenomenon so its impact also is global particularly in GHG observations. India faces drought during most ElNino years and photosynthesis activities are weak during this period and so CO2 enhancement occurs. I suggest authors to re-check your analysis in the case.

>We added “Indian Ocean Dipole” in L647 and L650. We checked the relationship of CO2 growth rate of Indian subcontinent sites and ENSO index many times because we also were surprise to see that in first time. We re-check the relationships after we got the CO2 data record in 30-50 years of Indian continent sites. As a side note, CO2 growth rate of few sites in Inland of Eurasia (Kazakhstan, Russia and China) also have anticorrelation with ENSO index.

Manuscript may be accepted for publication after addressing above comments. And GHG observations data at NTL and CLA should be made available to the researchers for further useful research.

>We are ready to open our observational data soon after publication.

Revised by authors: