Accepted Manuscript

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PII: S1352-2310(17)30803-8

DOI: [10.1016/j.atmosenv.2017.11.042](https://doi.org/10.1016/j.atmosenv.2017.11.042)

Reference: AEA 15699

To appear in: *Atmospheric Environment*

Received Date: 13 May 2017

Revised Date: 26 October 2017

Accepted Date: 26 November 2017

Please cite this article as: Guha, T., Tiwari, Y.K., Valsala, V., Lin, X., Ramonet, M., Mahajan, A., Datye, A., Kumar, K.R., What controls the atmospheric methane seasonal variability over India?, *Atmospheric Environment* (2017), doi: 10.1016/j.atmosenv.2017.11.042.

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The weaker CH_4 concentrations above Sinhagad (SNG) and Cape Rama (CRI) India during summer monsoon months JJAS may characterize a southern hemisphere origin. In order to verify this we compared the SNG and CRI CH_4 with that of the southern hemispheric values taken from Seychelles at Mahe Island (SEY). Noticeably the SEY CH_4 has similar seasonal variability as that of SNG and CRI, but with weaker amplitude probably because of its location over the open ocean. The in-phase seasonality between SNG, CRI and SEY thus suggest a possible southern hemisphere origin of CH_4 that is observed over the Indian peninsula during JJAS.

Figure: Model (LMDz) simulated CH_4 concentration compared with observations a) SNG (Sinhagad) and SEY (Seychelles), and b) CRI (Cape Rama) and SEY (Seychelles).

What controls the atmospheric methane seasonal variability over India?

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Abstract

Atmospheric CH₄ observations from two ground-based stations within Indian subcontinent, namely, Sinhagad (SNG) and Cape Rama station (CRI) showed a strong seasonality with a minima ($\sim 1800 \pm 20$ ppb) during southwest monsoon (SWM; i.e. June-September, JJAS) and a maxima (2000 ± 30 ppb) during northeast monsoon (NEM i.e. December-February, DJF) with a peak-to-peak seasonality close to 200 ppb. The Indian summer (winter) monsoon is characterized with strong southwesterly (northeasterly) winds of oceanic origin at the surface level and strong easterly (westerly) jet streams aloft. The monsoon dynamics has pronounced impact on CH₄ variability over India and is analyzed with winds, Lagrangian trajectories, and 3-dimensional distributions of CH₄ simulated by a general circulation model. The model simulations suggest a consistent annual vertical structure (mean and sub-seasonal uncertainty) of CH₄ over India with a stark contrast in concentration from summer to winter at surface levels (below 750 mb) in confirmation with what is identified by the ground-based observations. During SWM (NEM) the air with comparatively lower (higher) CH₄ concentrations from southern (northern) hemisphere reduces the CH₄ over India by 1814 ± 26 ppb (enhances by 1950 ± 51 ppb). The contribution of local fluxes to this seasonality appears to be albeit weak as the synthesized CH₄ fluxes (from EDGAR dataset) of the Indian peninsula itself show a peak in summer and a dip in winter. Similar property of CH₄ is also common to nearby oceanic region (i.e. over Arabian Sea, 1765 ± 10 ppb during summer) suggesting the role of monsoon dynamics as the controlling factor. Further the mixing and convection carries the CH₄ to the upper atmosphere and advect inward or outward aloft according the seasonal monsoon dynamics.

Keywords: Atmospheric CH₄ observations; Seasonal variation over India; CH₄ simulations

1. Introduction

The atmospheric methane (CH_4) is of chief concern as it is the second largest contributor to the global warming after carbon dioxide (CO_2), with a warming potential estimated to be $\sim 0.48 \pm 0.05 \text{ Wm}^{-2}$ (Fifth Assessment Report, IPCC, 2013). In addition to its global warming potential the CH_4 also reduces the oxidation chemistry of the atmosphere via its reaction with OH radicals (Kirschke et al., 2013). Atmospheric CH_4 was steadily increasing ($12 \pm 6 \text{ ppb yr}^{-1}$) in the 1980's. Thereafter the growth rate was slow in the 1990's, but again by 2007, rose to a growth rate of 8 ppb yr^{-1} (Dlugokencky et al., 2009).

The atmospheric CH_4 has both natural and anthropogenic origins. The wetland systems act as primary natural source of CH_4 (Cao et al., 1998) while rice paddies (Purkait et al., 2005; Khalil et al., 2008), livestock (Crutzen et al., 1986; Naqvi and Seijan, 2011), landfills (Chakraborty et al., 2011), combustion of fossil fuel, agricultural waste, biofuel (Howarth et al., 2011; Lelieveld et al., 1998), etc. acts as the dominant anthropogenic sources. The major source of CH_4 for Indian region is enteric fermentation of cattle, rice cultivation, waste water treatment etc (EDGAR HTAP database 2000-2010; http://edgar.jrc.ec.europa.eu/htap_v2/). The major sink of CH_4 in the atmosphere is via its reaction with OH radicals within the troposphere (Kirschke et al., 2013; Patra et al., 2011, Patra et al., 2014; Ghosh et al., 2015). Although the possible sources and sinks of CH_4 are well understood the accuracy of its global budget has large uncertainty especially at regional scales (Patra et al., 2016). Prima facie this difficulty is due to the distinct sources and sinks of CH_4 which vary widely with the seasons. The predicament is also escalated by the shortage of observations that capture the complex variations of CH_4 fluxes within a region (Patra et al., 2016).

Based on both top-down and bottom up approach, the annual flux of CH₄ was estimated to be 37.9 ± 3.6 and 38.5 ± 5.2 TgCyr⁻¹ from South Asian Region (Bangladesh, Bhutan, India, Nepal, Pakistan and Sri Lanka) (Patra et al., 2013). The discrepancy between estimated using both top-down and bottom up approach can be solved by expanding the atmospheric observation stations in South Asia (Patra et al., 2013). The atmospheric CH₄ variability over Indian subcontinent can be attributed to two major processes. The first is related to the CH₄ source itself. In India, the major anthropogenic sources of CH₄ includes livestock, rice paddies etc. An average CH₄ flux from Indian livestock was estimated to be 74.4 kg/hectare (Chhabra et al., 2013). Rice paddies are the major crop in India and its production increases following the southwest monsoon (hereafter SWM; from June-September; JJAS) due to the enhanced rain. This potentially increases the CH₄ emissions (Khalil et al., 2008). The second is atmospheric transport related; during SWM the lateral transport of air could bring inter-hemispheric CH₄ over the Indian region. The monsoon related deep convection over India could alleviate the surface CH₄ by transporting it to the mid/upper troposphere. The deep convection during SWM is an important factor in the trace gas dynamics over the Indian subcontinent (Ravi et al., 2013; Tiwari et al., 2014; Tania et al., 2015; Revadekar et al., 2016). This convection, in fact, is associated with a large scale upper level anticyclone over Tibetan plateau which forms at the beginning of SWM, propagates westward during latter part of the season and disappears by October-November (Krishnamurti and Balme, 1976; Hoskins and Rodwell, 1995; Hsu et al., 1999, Randel et al., 2010). Studies show that the Tibetan anticyclone could trap pollutants in mid/upper troposphere and mitigate them westward by the strong easterly jet streams (Randel et al., 2010; Schuck et al., 2010; Fadnavis et al., 2014). A similar dynamics to CH₄ can be expected at the upper atmosphere but its relation to surface observed CH₄ over India is not well understood.

Atmospheric Infrared Sounder (AIRS) satellite retrievals and model simulations showed an enrichment of CH₄ at mid-troposphere with a plume-like structure over South Asian region during JJAS (Xiong et al., 2009; Schuck et al., 2010). Arguably this enhancement could be due to the deep convective transport of surface CH₄ during SWM as detectable by data from trans-flight CH₄ measurement programs (CARIBIC; Schuck et al., 2009, 2010, 2012, CONTRAIL; Machida et al., 2008). The trans-flight measurements showed an enhancement of CH₄ concentrations at an altitude of 8-12km over a vast region of 50°E-80°E and south of 40°N. Schuck et al., (2010, 2012) showed that the CH₄ data from ground based monitoring station at Mauna Loa, Hawaii Islands is also comparable to that of the upper atmospheric (CARIBIC) observations. On contrary a station that is located over India (Cape Rama; 18.35° N, 73.75° E) showed a diminished CH₄ during SWM plausibly because the former station is out of the core Indian monsoon zone. This aspect, however, is not well explored in previous studies.

Aside from few observations of upper atmospheric CH₄, its surface monitoring over Indian subcontinent is sparse in order to decipher any relation between CH₄ variability and monsoon (Bhattacharya et al., 2009; Ganesan et al., 2013, 2015; Lin et al., 2015; Sreenivas et al., 2015). The seasonality of CH₄ found to be varying differently over different parts of India (Lin et al., 2015). For example a station at Darjeeling (Eastern Himalaya) captures episodes of higher CH₄ concentrations throughout the year (Ganesan et al., 2013). On the other hand during SWM the CH₄ over a station at Hanle (situated at northwestern Himalaya) captures higher values while stations at Pondichery and Andaman Islands (situated at southeast coast of peninsular India and Bay of Bengal, respectively) capture comparatively lower values (Lin et al., 2015). Such

complex spatiotemporal variability is attributed to seasonally varying CH_4 sources and monsoonal winds (Ganesan et al., 2013, 2015; Lin et al., 2015).

The column averaged CH_4 concentration (XCH_4) from SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) showed a spatiotemporal variation over Indian region (Kavitha and Nair, 2016a). The northern region captured seasonal peak in monsoon and post monsoon, whereas the peak was shifted to the end of October-December for the Southern peninsula region. This regional heterogeneity of XCH_4 is closely related with the livestock population, rice cultivation, wetland emissions etc. (Kavitha and Nair, 2016a). Upper tropospheric ($\sim 8 - 10\text{km}$) CH_4 from CARIBIC Aircraft measurement also showed seasonal high in monsoon, post monsoon and low in winter (Kavitha and Nair, 2016b). In comparison to both the column averaged and upper air CH_4 observation, near-surface observation showed contrasting seasonality where CH_4 peaks in winter and reaches its low in pre-monsoon and monsoon (Kavitha and Nair, 2016b). During monsoon convective activity uplifts CH_4 from boundary layer to upper troposphere and thus reduces the concentration at the surface. Convection, vertical transport and boundary layer height variation are identified as the possible factors for creating the contrast between near-surface and upper air observation of CH_4 (Kavitha and Nair, 2016b). Occasional high or low CH_4 concentrations over distinct parts of the atmosphere over Indian subcontinent were reported in several of the previous studies. However a linkage between them and its association with underlying monsoonal dynamics is largely unknown. In the current study we attempt to link the surface and upper air variability in atmospheric CH_4 to get a 3-dimensional distribution of it over Indian subcontinent

This work, for the first time, highlight the extreme contrasting origins of CH₄ arriving over a small subcontinent like India by the dramatic changes in the persisting seasonal atmospheric circulation. The contemporary observations of greenhouse gases are very limited and sparse in regions like India. Therefore one has to rely on models and satellite data to study the variability of greenhouse gases in such regions. In this view, the performance of the model is ultimately important. In this paper with the available observations we validated the model. The model can be a very good tool considering the accuracy it achieved when compared to two surface stations and airplane observations over India. None of the previous studies attempted and made a case that the model indeed are reliable, though with given uncertainty, and can be utilized for further study about CH₄ variability over India.

2. Methods

2.1. Surface CH₄ observations

The present study is based on the observation of atmospheric CH₄ concentration over 3 different sites located in the western India and the Southern Hemisphere (Figure 1). Along with the site locations the figure shows NCEP reanalysis of wind at surface (1000mb) and at upper level (200mbar) for January and July month representing two seasons. During July (representative for JJAS months), strong westerly / southwesterly wind prevails at the surface whereas at the upper troposphere (~200mb) strong easterly jet stream predominates; whereas in January (representative for DJF months) wind at the surface is northeasterly whereas at the upper troposphere strong westerly jet stream prevails. There are two greenhouse gas monitoring sites in western India; 1) Sinhagad (hereafter referred to as SNG) situated 200 km east of the Arabian Sea (18.35° N, 73.75° E) and located over the Western Ghats mountains at an elevation of 1600

m (asl); 2) Cape Rama (hereafter referred to as CRI), is a coastal site at Goa (15.08° N, 73.83° E, elevation = 50 m asl; Figure 1). Routine air samples at SNG site were collected from a 10m meteorological tower at a weekly interval from January, 2012 till December, 2013. The site has less vegetation coverage and the prevailing wind speed during the sampling time (i.e. noon) is comparatively low (of the day) ($0.5 - 1 \text{ m s}^{-1}$) for air sample to have an effect by the local sources and vegetation (Tiwari et al., 2014). Details of the air sampling method is given in Tiwari and Ravi Kumar, 2011, published in World Meteorological Organization (WMO) report No. 194 (Ravi Kumar et al., 2013).

Air samples collected from SNG site were brought back to Gas Chromatography (GC) lab at Indian Institute of Tropical Meteorology (IITM) for the analysis. The CH_4 concentration in the collected air samples was determined using Flame Ionization Detector (FID) detector of Thermo Trace Gas GC (Trace Gas Ultra, model no. KAV00338). The detail analytical procedure of the chromatographic run is given in Ravi Kumar et al., (2013). The measurement of CH_4 concentration in the air sample was calibrated to WMO standards. The GC analysis was calibrated for CH_4 measurement using International Standards provided by the WMO Central Calibration Laboratory (CCL) at the National Oceanic and Atmospheric Administration (NOAA)/Earth System Research Laboratory (ESRL)/Global Monitoring Division (GMD), Boulder, Colorado, USA. To understand the reproducibility in measurement, paired air samples were collected from SNG site and analyzed in the GC. The paired reproducibility was found to $\pm 5 \text{ ppb}$.

At CRI site, maintained by Commonwealth Scientific and Industrial Research Organization (CSIRO) Atmospheric Research GASLAB (Global Atmospheric Sampling

Laboratory), air sampling was going on since February, 1993 till January, 2013 with a few years of sampling gap during October, 2002 to July, 2009 (Francey et al., 1996; Bhattacharya et al., 2009). The observation obtained during July, 2009 till January, 2013 from CRI was considered here. Both the SNG and CRI site observations were compared with another station location in Northern Indian ocean (i.e. ~500km south of equator) for the sake of linking the large-scale monsoon dynamics and CH₄ variability over Indian subcontinent. The stations we chose are Seychelles at Mahe Island (4.67°S; 55.17°E; elevation=7m asl) (mentioned hereafter as SEY) maintained by NOAA/ESRL (WDCGG). The SEY site was operational since May, 1983 till today (WDCGG). In order to compare this site data with SNG and CRI, the observation obtained for the period of July, 2009 till December, 2013 was considered in the current study.

Both the SNG and CRI stations receive the seasonal reversal of wind pattern at surface level associated with both summer and winter monsoons. It is the primary meteorological feature of these sites (Tiwari et al., 2014). The study was focused mainly on SWM (hereafter referred to as JJAS) and winter monsoon i.e. December-February (hereafter referred to as DJF).

2.2. Model Simulations

The Indian subcontinent and its surrounding area are largely controlled by the monsoon meteorology both in terms of dynamics and composition of lower and upper troposphere trace gases. Thus in order to understand the effect of the Indian monsoon on the dynamics and dispersion of the atmospheric CH₄ at the surface and at the upper troposphere, the chemistry transport model LMDz (Lin et al., 2017) is utilized to simulate the CH₄ concentrations in the atmosphere. LMDz is general circulation model which is used here with a horizontal resolution of 0.5 x 0.66 latitude by longitude spanning 50–130°E and 0–55°N centered over India. Model

has 19 sigma coordinate layers in the vertical. The model solves the full dynamic equation for winds and then nudged towards European Centre for Medium-Range Weather Forecasts (ECMWF) analyzed meteorology with a relaxation time of 6 hours, in order to force transport to reproduce the observed large scale advection (Bousquet et al., 2005). Model uses advection scheme of Hourdin and Armengaud, 1999 and deep convection is parameterized using Tiedtke, 1989. Surface fluxes used to simulate the model are from different sources as anthropogenic emissions from EDGAR v4.2 FT2010 product (<http://edgar.jrc.ec.europa.eu>); wetland emissions from Kaplan et al., 2006; biomass burning emissions from GFED v3.0 product (<http://www.globalfiredata.org/data.html>); termite emissions from Sanderson, 1996; ocean emissions from Lambert and Schmidt, 1993; and soil uptake from Ridgwell et al., 1999. Model simulations are validated using various observations over the globe (Lin et al., 2016). Further details of the model and surface fluxes are described in Lin et. al., 2016.

Second model used in this study is Lagrangian particle dispersion model Flexpart. FLEXPART is an atmospheric transport model primarily designed to investigate the long-range and meso-scale dispersion of pollutants from point sources (Stohl et al., 2005, 2009). This model calculates the advection of multiple tracer particles either backward or forward in time using mean 3D winds and stochastic motions from the turbulence fluxes. It introduces discrete inert particles at each receptor location and moves them backward/forward in time in the model domain using prescribed meteorological field. Three hourly meteorological field data from the National Center for Environmental Prediction Global Forecasting System (NCEP) (<http://www.nco.ncep.noaa.gov/pmb/products/gfs/>) archived at $0.5^\circ \times 0.5^\circ$ grid resolution is used as inputs to FLEXPART simulations. In this study, backward simulations from both SNG and

CRI sites were made every 3 hours interval and are used to understand long range transport of air masses at surface monitoring sites. The starting period of the back-trajectory analysis was fixed on the 15th of the month. Twenty thousand particles are released from a 10 m height at the receptor location at 12:00 h UTC (measurement site) and FLEXPART calculates a 4D (space and time) response function (sensitivity) to emission input. 15-day backtrajectory was initiated at mid of the month during July and January. 15 days back flow is a representative of the characterize transport for the respective month. Detailed model simulations over surface sites in India are given in Tiwari et al., 2013, 2014.

2.3. Aircraft CH₄ observations

The CARIBIC system (<http://www.caribic-atmospheric.com>), loaded onto Lufthansa Airlines measures CO₂, CH₄, O₃, CO etc. and aerosol at an altitude of around 8 - 12.5 km (Schuck et al., 2009, 2012). Air samples are collected onboard using the Triggered Retrospective Air Collector (Brenninkmeijer et al., 2007). Post-flight concentration analysis of the collected trace gases are carried out using Gas Chromatography-Flame Ionization Detector (GC-FID) system at Max Planck Institute of Chemistry, Germany and the precision obtained for CH₄ measurement is 0.17% (Schuck et al., 2009). The air sampling along Frankfurt (FRA) to Chennai (MAA) route was initiated in April 2008 (Schuck et al., 2012), and continued till March 2012.

3. Result and Discussions

3.1. Seasonal variation of CH₄

The atmospheric CH₄ at SNG and CRI has a seasonality of ~200 ppb (Figure 2a, b). CH₄ concentration reaches its minima during the summer monsoon (i.e. during JJAS); whereas it

reaches its maxima during the winter monsoon (i.e. during DJF). During JJAS the CH₄ concentrations at SNG was minimum about 1821±21 ppb and 1818±21 ppb for years 2012 and 2013, respectively; whereas at CRI the minimum is about 1781±18 ppb, 1792±11 ppb and 1795±22 ppb years 2010, 2011 and 2012, respectively. On the other hand during DJF at SNG a maximum is observed about 1882±54 ppb and 2015±32 ppb for years 2012 and 2013, respectively; whereas at CRI the maximum is about 1910±30 ppb, 1922±28 ppb and 1927±20 ppb for years 2010, 2011 and 2012, respectively (see also Table 1). Here the uncertainty depicts the standard deviation of daily CH₄ with respect to the monthly seasonal average. In comparison to SNG the CRI is located at the coast and the site is comparably free from vegetation and local anthropogenic influences. Thus CRI captures lower magnitude of CH₄ concentration in comparison with SNG (Tiwari et al., 2014).

The lower CH₄ concentrations above SNG and CRI during JJAS may characterize a southern hemisphere origin. In order to verify this we compared the SNG and CRI CH₄ with that of the observations taken from SEY (see method for details). Noticeably the SEY CH₄ has similar seasonal variability as that of SNG and CRI, but with weaker amplitude probably because of its location over the open ocean. The in-phase seasonality between SNG, CRI and SEY thus suggest a possible southern hemisphere origin of CH₄ that is observed over the Indian peninsula during JJAS. The contribution of local fluxes to this seasonality appears to be insignificant as the synthesized CH₄ fluxes (EDGAR) of the Indian peninsula itself show a peak in summer and a dip in winter (Lin et al., 2017). Nevertheless, during the JJAS, the CH₄ at SNG, CRI, SEY all collectively have similar concentrations implying that the air-mass originated from southern hemisphere is visiting the sites.

Backward trajectories of particles initialized from SNG and CRI in summer monsoon season indicates transport of air mass from the southern hemisphere to the Indian subcontinent. The trajectories are also passing through the station SEY (Figure 3). Therefore the southern hemispheric CH_4 arrive SNG and CRI during summer monsoon explaining the similar concentration of CH_4 at all three sites examined here. During the winter monsoon, on the other hand, the trajectories indicate continental origin of CH_4 at SNG and CRI (see also Tiwari et al, 2014).

3.2. CH_4 variability from model simulations

The LMDZ simulated CH_4 concentration at SNG and CRI found to match exceptionally well with the ground-based observations (Figure 2a, 2b). To quantitatively understand the consistency between observation and model simulations the root mean square difference (RMSD) between them was calculated for each season. The RMSD was 68.7 (9.6) ppb during summer monsoon and 114.3 (33.8) ppb during winter monsoon for SNG and CRI, respectively. Both the ground-based observation and model was found to be highly consistent with each other with a low RMSD for JJAS compared to DJF. The LMDz simulated CH_4 at upper level also showed close match with CARIBIC observations (Figure 4). The CARIBIC flight track (Figure 4a) facilitates the understanding of model offset over both the Arabian Sea and the Indian subcontinent. LMDz model simulated CH_4 was sub-sampled along the CARIBIC flight track for individual days. Model offset was calculated by subtracting the CARIBIC observations from sub-sampled model simulated CH_4 . The spatial pattern of model offset along longitude showed large variation between different months in winter over ocean sector whereas near to Indian subcontinent and nearby region, the consistent offset with an average of 12 ± 4 ppb was found for

winter of 2011 (Figure 4b). The landing of CARIBIC flight over Chennai, India provided the vertical profile of model offset over Indian subcontinent. In vertical profile model offset was found to be 7 ± 4 ppb (Figure 4c). In summary, the model offset indicates that during winter model is over-estimating CH_4 concentration for Indian subcontinent and its surrounding region. The validation of LMDz model with ground based, and aircraft based observation confirms that the model simulations are faithful with respect to the available observations of CH_4 from Indian subcontinent analyzed here.

The consistency between observation and model allowed us to portray a 3-dimensional structure of atmospheric CH_4 distribution during JJAS and DJF over Indian subcontinent and its surrounding region. During southwest monsoon the transport of southern hemispheric clean air at surface causes lower concentration. Upon arrival at the continent it gets mixed with the surface emitted CH_4 . Then the deep convection over continent carries the ground level higher emission to the upper atmosphere. At the upper atmosphere there is a presence of strong easterly jet stream which carries the higher CH_4 concentration from continent to the nearby ocean i.e. over Arabian Sea. Thus there was higher CH_4 over Arabian Sea during southwest monsoon. During winter, the observation over continent is largely affected by the local emission at the surface. Then effect of surface emission decreases with increasing altitude. The decrease is more prominent over land than over ocean.

3.2.1 Vertical profile of atmospheric CH_4 over ocean and continent

The mean vertical profile of CH_4 from a point over the central India (78°E , 20°N) and over a nearby oceanic region (i.e. over Arabian Sea, 65°E , 17°N) was constructed based on the model outputs. The mean was calculated as monthly mean of 9-years (2006 till 2014) of model

simulations. An uncertainty to this mean at each level was also calculated as the standard deviations of daily CH₄ data for individual months from all years. In order make sure the standard deviation represent for the synoptic variability we have removed first two harmonic from each year before calculating the spread.

Major contrast between winter and summer profiles of CH₄ over Indian subcontinents are noticed below 750 mb (Figure 5). During winter the surface CH₄ over the continent was relative higher (1950±51 ppb) compared to the upper atmosphere (1814±26; above 750 mb). During summer, on the other hand, CH₄ appears to be well mixed over the continent possibly due to the summer time convections. This property is also observed over the nearby oceanic region but further lower concentrations of CH₄ at surface level (1765±10 ppb) as compared to the land possibly due to the transport of more ‘fresh’ air from the southern hemisphere to the Arabian Sea.

Above 750 mb, the concentration of CH₄ over continent as well as over the ocean appears to be similar throughout the year with slight deviation between summer and winter. This mean structure varies slightly by uncertainty contributed by the synoptic variability. Such fine scale variability could either be due to variability in atmospheric circulation or due to variability in fluxes. However there is no precise estimate of CH₄ fluxes at higher resolution to ascertain the role of transport versus fluxes in the uncertainty of the mean shown here.

3.2.2 Spatial distribution of atmospheric CH₄

In above section, vertical profiles over the land and the ocean provide the glimpses of structure of CH_4 over India as seen from winter and summer. To elaborate a three dimensional view of this vertical distribution of CH_4 we examined zonal average of LMDz simulated CH_4 concentration from 60E-75E and displayed as a meridional section from 59S- 22N. The sections are produced for both winter (Figure 6a) and summer (Figure 6b) separately. The inter-hemispheric gradient of CH_4 is well maintained at about equator during winter. The contours show that over the continent the surface CH_4 is somewhat ‘trapped’ in the lower atmosphere and subsidence from aloft, a typical feature of winter monsoon circulation.

A conspicuous nature of the CH_4 vertical profiles over the continent during summer monsoon is the vertical homogenization of concentrations by the convection (Figure 6b). The upper atmosphere experiences an intrusion of CH_4 concentrations from land to ocean. This is typical feature of a Hadley cell circulation established during JJAS over Indian sub-continent.

4. Conclusions

CH_4 observations over Indian subcontinent show interesting differences from other tropical sites (eg. Mauna Loa, Hawaii Islands, Shuck et al., 2010). Over Indian subcontinent the CH_4 concentrations appear to be influenced dominantly by the transport and convection rather than the fluxes which itself show a peak in summer following the rain and agricultural activities and a dip in winter (Lin et al., 2016). The summer convection, winter subsidence and lateral transport by monsoonal reversal all contribute to explain the CH_4 variability over India.

It appears that there is a difference in seasonality between the surface and upper atmospheric CH₄ over Indian subcontinent. The surface observations show that there is a dip in CH₄ during JJAS and a hike in DJF due to the lateral transport. On the other hand at the upper atmosphere the JJAS CH₄ is high compared to the DJF (Schuck et al., 2010, Xiong et al., 2009). The satellite observations are in fact capturing this seasonality in the upper atmosphere because the convection carries the surface fluxes to the upper atmosphere during JJAS. While the surface observations are influenced by the lateral transport and air-mass visiting from the southern hemisphere.

Atmospheric CH₄ observation at ground level showed a strong seasonality with minima in southwest monsoon and maxima in winter. Although the removal of CH₄ by higher OH radical is expected to play a role in southwest monsoon, the effect in the transport of southern hemisphere '*clean*' air on the CH₄ variability is not ignorable. In fact the visit of '*clean*' oceanic air and sink due to elevated OH radicals may contribute to the dilution of CH₄ over surface during JJAS.

We also quantified the effect of OH on CH₄ loss through oxidation processes during monsoon and winter. OH oxidation is the main loss process for CH₄ in the atmosphere ($k = 1.85 \times 10^{-12} e^{(-1690/T)}$; Atkinson et al., 2004). In the winter season, considering the background OH concentrations above the Indian subcontinent, the CH₄ lifetime computes to about approximately 1430 days. However, due to an increase OH concentrations in the monsoon season, the CH₄ lifetime is about 782 days, which is about half as much as during winter. To cause a reduction of about 130 ppbv (Figure 2) from average winter concentrations to average monsoon concentrations, it would take about 54 days during the monsoon season. This result indicates that

due to the long lifetime of CH_4 against oxidation, the large seasonal difference would not be controlled by OH oxidation, but rather is indicative of transport of different air masses.

The CO_2 concentrations in the atmosphere over Indian subcontinent are rather influenced by the net ecosystem exchange and the signals related to transport are obscured (Ravi et al., 2016). On the other hand the influence of transport and convection may be separable from the CH_4 variability in comparison with those induced by the surface fluxes. The CH_4 could advect in and out of the Indian subcontinent by the intermittent variability of monsoonal circulation within a season (Goswami et al., 2006). This should be examined in order to see whether the CH_4 fluctuations by monsoonal dynamics feedback any radiative value to the convection over India (Goswami et al., 2006).

Although the Indian monsoon and convections over Indian and adjacent Asian regions are well understood and in this context it may not be surprising to know that the associated CH_4 dynamics also behaving to the tune of the monsoon. However, there are important scientific questions exist behind this scenario as follows. CH_4 is much more active (20 times or more) than CO_2 in the radiative heat budget. Therefore the role of the CH_4 variability in the local monsoon radiation balance needs to be understood. Without background knowledge in CH_4 variation during monsoon one cannot achieve this target. This paper is the first and important step towards this goal. The results should be viewed with this spirit that the atmospheric structure of CH_4 during winter and summer can be extremely different over India irrespective of its sources, though uncertainty looms over this.

Acknowledgements

The authors would like to express thanks to CARIBIC project for providing upper troposphere methane data in Frankfurt to Chennai flight. We express our gratitude to Carl A. M. Brenninkmeijer and Armin Rauthe-Schoch from Max Planck Institute for Mainz Germany, for their helpful comments and discussion. We would like to thank Ray Langenfeld and Marcel Vanderschot from CSIRO Australia for sharing Cape Rama data. We are thankful to the Ministry of Earth Sciences Govt. of India for supporting research through Indian Institute of Tropical Meteorology Pune India.

References:

- 1) Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J., and Troe, J., 2004. Evaluated kinetic and photochemical data for atmospheric chemistry: Volume I - gas phase reactions of Ox, HOx, NOx and SOx species, *Atmos. Chem. Phys.*, 4, 1461-1738, 10.5194/acp-4-1461-2004, 2004.
- 2) Bhattacharya, S.K., Borole, D. V., Francy, R. J., Allison, C. E., Steele, L. P., Krummel, P., Langenfelds, R., Masarie, K. A., Tiwari, Y. K., Patra, P. K., 2009. Trace gases and CO₂ isotope records from Cabo de Rama, India. *Curr Sci.* 97(9), 1336–1344.
- 3) Bousquet, P., Hauglustaine, D. A., Peylin, P., Carouge, C., Ciais P., 2005. Two decades of OH variability as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform, *Atmos. Chem. Phys. Disc.*, 5, 1679–1737.
- 4) Brenninkmeijer, C.A.M., Crutzen, P., Boumard, F., Dauer, T., Dix, B., Ebinghaus, R., Filippi, D., Fischer, H., Franke, H., Frie, U., Heintzenberg, J., Helleis, F., Hermann, M.,

- Kock, H.H., Koeppel, C., Lelieveld, J., Leuenberger, M., Martinsson, B.G., et al., 2007. Civil Aircraft for the regular investigation of the atmosphere based on an instrumented container: the new CARIBIC system. *Atmos. Chem. Phys.* 7 (18), 4953e4976. <http://dx.doi.org/10.5194/acp-7-4953-2007>.
- 5) Cao, M., Gregson, K., Marshall, S., 1998. Global methane emission from wetlands and its sensitivity to climate change. *Atmos. Environ.* 32 (19), 3293e3299. [http://dx.doi.org/10.1016/S1352-2310\(98\)00105-8](http://dx.doi.org/10.1016/S1352-2310(98)00105-8).
- 6) Chakraborty, M., Sharma, C., Pandey, J., Singh, N., Gupta, P.K., 2011. Methane emission estimation from landfills in Delhi: a comparative assessment of different methodologies. *Atmos. Environ.* 45 (39), 7135e7142. <http://dx.doi.org/10.1016/j.atmosenv.2011.09.015>.
- 7) Chhabra, A., Manjunath, K. R., Panigrahy, S., Parihar J. S., 2013. Greenhouse gas emission from Indian livestock. *Climate Change*. (117), 329-344.
- 8) Ciais, P., Sabine, C., Bala, G., Bopp, L., Brovkin, V., Canadell, J., Chhabra, A., DeFries, R., Galloway, J., Heimann, M., Jones, C., Le Quéré, C., Myneni, R.B., Piao, S., Thornton, P., 2013. Carbon and Other Biogeochemical Cycles. 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., Qin, D. Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex V., Midgley, P. M. (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- 9) Dlugokencky, E.J., Bruhwiler, L., White, J. W. C., Emmons, L. K., Novelli, P. C., Montzka, S. A., Masarie, K. A., Lang, P. M., Crotwell, A. M., Miller, J. B., Gatti, L. V.,

2009. Observational constraints on recent increases in the atmospheric CH₄ burden. *Geophys. Res. Lett.* 36(18803), doi:10.1029/2009GL039780.
- 10) Fadnavis, S., Schultz, M. G., Semeniuk, K., Mahajan, A. S., Pozzoli, S., Sonbawne, S., Ghude, S.D., Kiefer, M., Eckert E., 2014. Trends in peroxyacetyl nitrate (PAN) in the upper troposphere and lower stratosphere over southern Asia during the summer monsoon season: regional impacts. *Atmos. Chem. Phys.* 14, 12725–12743.
- 11) Francey, R.J., L.P. Steele, R.L. Langenfelds, M.P. Lucarelli, C.E. Allison, D.J. Beardsmore, S.A. Coram, N. Derek, F.R. de Silva, D.M. Etheridge, P.J. Fraser, R.J. Henry, B. Turner, E.D. Welch, D.A. Spencer and L.N. Cooper. Global Atmospheric Sampling Laboratory (GASLAB): supporting and extending the Cape Grim trace gas programs. *Baseline Atmospheric Program (Australia) 1993*, edited by R.J. Francey, A.L. Dick and N. Derek, pp 8 - 29, Bureau of Meteorology and CSIRO Division of Atmospheric Research, Melbourne, Australia, 1996.
- 12) Ganesan, A.L., Chatterjee, A., Prinn, R. G., Harth, C. M., Salameh, P. K., Manning, A. J., Hall, B. D., Mühle, J., Meredith, L. K., Weiss, R. F., O'Doherty, S., Young, D., 2013. The variability of methane, nitrous oxide and sulfur hexafluoride in Northeast India. *Atmos. Chem. Phys.* 13, 10633–10644.
- 13) Ganesan, A. L., Manning, A. J., Grant, A., Young, D., Oram, D. E., Sturges, W. T., Moncrieff, J. B., O'Doherty, S., 2015. Quantifying methane and nitrous oxide emissions from the UK and Ireland using a national-scale monitoring network.
- 14) Ghosh, A., Patra, P. K., Ishijima, K., Umezawa, T., Ito, A., Etheridge, D. M., Sugawara, S., Kawamura, K., Miller, J. B., Dlugokencky, E. J., Krummel, P. B., Fraser, P. J., Steele, L. P., Langenfelds, R. L., Trudinger, C. M., White, J. M. C., Vaughn, B., Saeki, T. Aoki,

- S., Nakazawa T., 2015. Variations in global methane sources and sinks during 1910–2010. Atmos. Chem. Phys. 2595-2612, doi:10.5194/acp-15-2595-2015.
- 15) Hoskins, B. J. and Rodwell, M. J., 1995. A Model of the Asian Summer Monsoon. Part I: The Global Scale., J. Atmos. Sci., 52, 1329– 1340, doi:10.1175/1520-0469(1995)052.
- 16) Hourdin, F., Armengaud, A., 1999. Test of a hierarchy of finite-volume schemes for transport of trace species in an atmospheric general circulation model, Mon. Weather Rev., 127, 822– 837.
- 17) Howarth, R. W., Santoro, R., Ingraffea, A., 2011. Methane and the greenhouse-gas footprint of natural gas from shale formations. Climate Change, 106 (4), 679-690. DOI: 10.1007/s10584-011-0061-5
- 18) Hsu, H.-H., Terng, C.T., Chen, C.T., 1999 Evolution of Large- Scale Circulation and Heating during the First Transition of Asian Summer Monsoon., J. Climate, 12, 793–810, doi:10.1175/1520-0442(1999)012.
- 19) IPCC: IPCC: Climate Change 2013. The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, 2013.
- 20) IPCC: Climate Change 2014. Impacts, Adaptation, and Vulnerability. Part A: Global and Sectoral Aspects. Contribution of Working Group II to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Field, C. B., Barros, V. R., Dokken, D. J., Mach, K. J., Mastrandrea, M. D., Bilir, T. E., Chatterjee, M., Ebi, K. L., Estrada, Y. O., Genova, R. C., Girma, B., Kissel, E. S., Levy, A. N., MacCracken, S., Mastrandrea, P. R., and White, L. L., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2014.

- 21) Kaplan, J. O., Folberth, G. and Hauglustaine, D. A.: Role of methane and biogenic volatile organic compound sources in late glacial and Holocene fluctuations of atmospheric methane concentrations, *Global Biogeochem. Cycles*, 20(2), GB2016, doi:10.1029/2005GB002590, 2006.
- 22) Kavitha, M., Nair, P.R., 2016a. Region-dependent seasonal pattern of methane over Indian region as observed by SCIAMACHY. *Atmos. Environ.* 131, 316e325. <http://dx.doi.org/10.1016/j.atmosenv.2016.02.008>.
- 23) Kavitha, M., Nair, P.R., 2016b Non-homogeneous vertical distribution of methane over Indian region using surface, aircraft and satellite based data. *Atmos. Environ.* 141, 174-185.
- 24) Keeling, C. D., Whorf, T. P., Wahlen, M., Van der Plicht, J., 1995. Interannual extremes in the rate of rise of atmospheric carbon dioxide since 1980, *Nature*, 375, 666–670.
- 25) Khail M. A. K., Shearer, M. J., Rasmussen, R. A., Duan, C., Ren, L., 2008. Production, oxidation, and emissions of methane from rice fields in China. *J. Geophys. Res.* 113 (G00A04), doi:10.1029/2007JG000461.
- 26) Kirschke et al.: Three decades of global methane sources and sinks, *Nature Geoscience*, DOI:10.1038/NGEO1995, 2013.
- 27) Krishnamurti, T. N. and Bhalme, H. N.: Oscillations of a Monsoon System. Part I. Observational Aspects., *J. Atmos. Sci.*, 33, 1937– 1954, doi:10.1175/1520-0469(1976)033, 1976.
- 28) Lambert, G. and Schmidt, S.: Reevaluation of the oceanic flux of methane: Uncertainties

and long term variations, *Chemosphere*, 26(1–4), 579–589,
doi:[http://dx.doi.org/10.1016/0045-6535\(93\)90443-9](http://dx.doi.org/10.1016/0045-6535(93)90443-9), 1993.

29) Lin X., Ciais P., Bousquet P., Ramonet M., Yin Y., Balkanski Y., Cozic A., Delmotte M.,
Evangelidou N., Indira N. K., Locatelli R., Peng S., Piao S., Saunio M., Swathi P. S.,
Wang R., Yver-Kwok C., Tiwari Y. K., and Zhou L.: Simulating CH₄ and CO₂ over South
and East Asia using the zoomed chemistry transport model LMDzINCA, *Atmos. Chem.*
Phys. Discuss., doi:10.5194/acp-2016-1056, 2017.

30) Machida, T Worldwide measurements of atmospheric CO₂ and other trace gas species
using commercial airlines, *J. Atmos. Oceanic Technol.*, 25(10), 1744–1754,
doi:10.1175/2008JTECHA1082.1.

31) Myhre, G., D. Shindell, F. M. Bréon, W. Collins, J. Fuglestad, J. Huang, D. Koch, J.-F.
Lamarque, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura and
H. Zhang, 2013: Anthropogenic and Natural Radiative Forcing. 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.

32) Naqvi, S., M. K., Seijan, V., 2011. Global climate change: role of livestock. *Asian J.*
Agric. Sci. 31 (1), 19e25.

33) Patra, P. K., Canadell, J. G., Houghton, R. A., Piao, S. L., Oh, N.-H., Ciais, P., Manjunath,
K. R., Chhabra, A., Wang, T., Bhattacharya, T., Bousquet, P., Hartman, J., Ito, A.,

- Mayorga, E., Niwa, Y., Raymond, P. A., Sarma, V. V. S. S., Lasco, R., 2013. The carbon budget of South Asia. *Biogeosciences*, 10, 513-527.
- 34) Patra, P. K., Krol, M. C., Montzka, S. A., Arnold, T., Atlas, E. L., Lintner, B. R., Stephens, B. B., Xiang, B., Elkins J. W., Fraser, P. J., Ghosh, A., Hints, E. J., Hurst, D. F., Ishijima, K., Krummel, P. B., Miller, B. R., Miyazaki, K., Moore, F. L., Mühle, J., O'Doherty, S., Prinn, R. G., Steele, L. P., Takigawa, M., Wang, H. J., Weiss, R. F., Wofsy, S. C., Young, D., 2014. Observational evidence for interhemispheric hydroxyl parity, *Nature*, 513, 219-223.
- 35) Patra P., Saeki T., Dlugokencky, E. J., Ishijima, K., Umezawa, T., Ito, A., Aoki, S., Morimoto, S., Kort, E. A., Crotwell A., 2016. Regional Methane emission estimation based on observed atmospheric concentrations (2002-2012), *J. Of Meteorological Society of Japan*, 94(1), 85-107.
- 36) Purkait N. N., Sengupta M. K., Sanghamitra, De, Chakraborty D. K. Methane emission from the rice fields of West Bengal over decades. *Indian J. Radio Spac Phys.* 34, 255-263.
- 37) Randel, W. J., Park, M., Emmons, L., Kinnison, D., Bernath, P., Walker, K. A., Boone, C., and Pumphrey, H. 2010. Asian monsoon transport of pollution to the stratosphere. *Science*, 328, 611–613, doi:10.1126/science.1182274.
- 38) Ravi Kumar K., Yogesh K. Tiwari, Vinu Valsala, Raghu Murtugudde, 2014, On understanding of land-ocean CO₂ contrast over Bay of Bengal: A case study during 2009 summer monsoon, *Environmental Science and Pollution Research*, Volume 21, Issue 7, April 2014, Page 5066-5075, DOI: 10.1007/s11356-013-2386-2.
- 39) Ravi Kumar, K., Valsala, V., Tiwari, Y. K., Revadekar, J. V., Pillai, P., Chakraborty, S., Murtugudde, R., 2016. Intra-seasonal variability of atmospheric CO₂ concentrations over

India during summer monsoons. *Atmos. Environ.* 142 (2016) 229e237, DOI: 10.1016/j.atmosenv.2016.07.023.

40) Revadekar J. V., Ravi Kumar K., Tiwari Y. K. and Valsala V. Variability of AIRS CO₂ during active and break phases of Indian summer monsoon, *Sci. of Total Envir.* 541;1200-1207, 2016.

41) Ridgwell, A. J., Marshall, S. J. and Gregson, K.: Consumption of atmospheric methane by soils: A process-based model, *Global Biogeochem. Cycles*, 13(1), 59–70, doi:10.1029/1998GB900004, 1999.

42) Sanderson, M. G.: Biomass of termites and their emissions of methane and carbon dioxide: A global database, *Global Biogeochem. Cycles*, 10(4), 543–557, doi:10.1029/96GB01893, 1996.

43) Schuck, T. J., Brenninkmeijer, C. A. M., Slemr, F., Xueref-Remy, I., and Zahn, A.: Greenhouse gas analysis of air samples collected onboard the CARIBIC passenger aircraft, *Atmos. Meas. Tech.*, 2, 449–464, 2009.

44) Schuck, T. J., Brenninkmeijer, C. A. M., Baker A. K., Slemr F., von Velthoven, P. F. J., and Zahn A. Greenhouse gas relationships in the Indian summer monsoon plume measured by the CARIBIC passenger aircraft, *Atmos. Chem. Phys.*, 10(8), 3965–3984, doi:10.5194/acp-10-3965-2010 (2010).

45) Schuck, T. J., Ishijima K., Patra P. K., Baker A., Machida T., Matsueda H., Sawa, Y., Umezawa, T., Brenninkmeijer, C. A. M., and Lelieveld, J. Distribution of methane in the tropical upper troposphere measured by CARIBIC and CONTRAIL aircraft. *J. Geophys. Res.* 117, D19304 (2012).

- 46) Stohl, A., Seibert, P., Arduini, J., Eckhardt, S., Fraser, P., Grealley, B. R., et al. 2009. An analytical inversion method for determining regional and global emissions of greenhouse gases: sensitivity studies and application to halocarbons. *Atmos. Chem. Phys.* 9, 1597–620.
- 47) Tania, G., Ghosh, P., 2015, Diurnal and seasonal variation of mixing ratio and $\delta^{13}\text{C}$ of air- CO_2 observed at an urban station Bangalore, India, *Envir Sci & Pollut Research*, doi: 10.1007/s11356-014-3530-3.
- 48) Tiwari, Y. K., Ravi Kumar, K., 2011. Glass flask air sample analysis through gas chromatography in India: implications for constraining CO_2 surface fluxes, WMO/GAW Report No. 194, WMO/TD-No.1553, April 2011.
- 49) Tiwari, Y. K., Valsala, Vinu, Vellore, Ramesh K., Ravi Kumar, K., 2013. Effectiveness of surface monitoring stations in representing regional CO_2 emissions in India, *Clim. Res.*, Vol. 56: 121–129, 2013, doi: 10.3354/cr01149
- 50) Tiwari, Y. K., Vellore, R. K., Ravi Kumar, K., Schoot, M. V. D., Cho, C., 2014. Influence of monsoons on atmospheric CO_2 spatial variability and ground-based monitoring over India, *Science of The Total Environment*. 490 (15), 570–578, <http://dx.doi.org/10.1016/j.scitotenv.2014.05.045>.
- 51) Tiedtke, M., 1989. A comprehensive mass flux scheme for cumulus parameterization in large-scale models, *Mon. Weather Rev.*, 117, 1179–1800.
- 52) Xiong, X., Houweling, S., Wei, J., Maddy, E., Sun, F., Barnett, C.: Methane plume over south Asia during the monsoon season: satellite observation and model simulation, *Atmos. Chem. Phys.*, 9, 783-794, 2009.

Table 1: Seasonal average of Atmospheric CH₄ concentration at Sinhagad (SNG) and Cape Rama (CRI) during JJAS and DJF of 2010 - 2013

	SNG		CRI	
	JJAS	DJF	JJAS	DJF
2010	-	-	1781±18	1910±30
2011	-	-	1792±11	1922±28
2012	1821±21	1882±54	1795±22	1927±20
2013	1818±21	2015±32	-	-

Figure 1: NOAA NCEP-derived monthly mean u-wind at the surface (1000 mb) and at upper troposphere (200 mb) during July (left panel) and January (right panel). Arrows indicate wind direction. Stations SNG (Singahad, India), CRI (Cape Rama, India), and SEY (Seychelles) are marked in black rectangles.

Figure 2: Measured and LMDz model simulated CH_4 concentration at a) SNG (Sinhagad) and SEY (Seychelles), and b) CRI (Cape Rama) and SEY (Seychelles).

Figure 3: Lagrangian back-trajectories of particles simulated by FLEXPART starting from station SNG (Sinhagad) and CRI (Cape Rama) during south west monsoon month July (left panel) and winter month January (right panel). Colors indicate position of particles in the vertical (m).

Figure 4: (a) CARIBIC flight track from Frankfurt to Chennai on 16th Jan., 2012, rectangular boxes show oceanic sector and continental sector; (b) Longitudinal variation of CH₄ concentration (model minus observed) over oceanic sector, vertical black line represents continental boundary; and (c) Vertical profile of CH₄ concentration (model minus observed) over continental sector.

Figure 5: Model simulated vertical profile of CH_4 over (a) Arabian Sea and (b) Indian subcontinent during summer (Jul, Aug) and winter (Dec, Jan) months.

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648 Figure 6: Vertical profile of model simulated CH₄ concentration averaged over longitude
 649 bands 60°E-80°E and during 2006 to 2013 a) Jan and b) Jul. Contour lines indicates model
 650 CH₄ concentration values.

- A Summer minima and winter maxima of CH₄ at surface is observed over India
- A seasonality of ~200 ppb is linked with the atmospheric circulation and convection
- Seasonality of CH₄ is confined to below 750 mb with uniform profile of CH₄ aloft
- The low-level convergence, uplift & upper level divergence shapes CH₄ over India