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Non-homogeneous vertical distribution of methane over Indian region using surface, aircraft and satellite based data

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HIGHLIGHTS

- Non-homogeneous distribution of methane from surface to upper troposphere.
- Convective activity plays a role in the vertical distribution of methane.
- Latitudinal difference in seasonal pattern of the upper troposphere methane.
- Long range transport effective in the upper troposphere.
- Boundary layer height controls near-surface methane level.

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ABSTRACT

The upper tropospheric methane (UCH_4) data from Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container (CARIBIC), the column average mixing ratio of methane (XCH_4) from SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) and near-surface methane at two locations- Cape Rama, Goa, a Commonwealth Scientific Industrial Research Organization (CSIRO) network station and Ahmedabad (23.03°N; 72.45°E) were analysed to understand vertical inhomogenities in methane mixing ratio and the seasonal changes in the latitude sector 13°–24°N over India. XCH_4 and UCH_4 were found to follow more or less similar pattern over all the three latitude sectors, with the peak occurring in July–August, and minimum in late winter. The seasonal amplitude in XCH_4 is less at low latitude sector (~64 ppbv) compared to that of high latitudes (~101 ppbv at 18°–22°N and 88 ppbv at 22°–24°N). On the other hand, the near surface methane shows opposite pattern peaking in winter attaining low in monsoon. During monsoon when methane sources are active at the surface, $XCH_4 > UCH_4$ and during other seasons $UCH_4 > XCH_4$ indicating presence of high altitude layers. This analysis revealed non-homogeneous distribution of methane in the troposphere indicative of stratified layers. The role of convective activity, boundary layer meteorology and long-range transport in controlling the seasonal changes in the vertical distribution of methane are examined in this study.

1. Introduction

Rising concern over the increase in anthropogenic greenhouse gas emissions and their dangerous consequences on global climate has fuelled systematic monitoring of these gases all over the globe. Methane (CH_4) is the second-most important anthropogenic greenhouse gas after carbon dioxide (CO_2) in terms of net radiative forcing (Stocker et al., 2013), contributing 15% of the enhanced greenhouse effect. Also, it is the most abundant reactive green house gas in the atmosphere, playing vital roles in the energy balance and chemistry of the troposphere and stratosphere regions of the atmosphere. CH_4 is emitted from a wide variety of natural and anthropogenic sources and classified as biogenic, thermogenic and pyrogenic. Biogenic process involves methanogenic action on organic matter in natural wetlands, water flooded rice paddies, landfills and in the ducts of ruminant animals, etc. (Moss et al., 2000; Nosalewicz et al., 2011). Emission associated to fossil fuel mining, processing, transportation and distribution comes under thermogenic sources of CH_4. Pyrogenic CH_4 is produced as a result

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of incomplete combustion of organic material like burning of agricultural wastes, forests, biofuels, etc. (Lelieveld et al., 1998; Worden et al., 2013).

About 90% of CH_4 in the atmosphere breaks down by reacting with hydroxyl radical (OH) and the remaining is lost in biological and stratospheric oxidation (Portmann et al., 2012). The reaction between CH_4 and OH, in the presence of high levels of nitrogen oxides (NOx) produces species like formaldehyde (HCHO), carbon monoxide (CO), ozone (O_3), H_2O etc. through a series of complex reactions (Levy, 1971) and thus controls the amount of OH in the atmosphere. The stratospheric CH_4 that is intruded from troposphere follows different chemistry. CH_4 also plays vital role in stratospheric O_3 chemistry. Oxidation of CH_4 in stratosphere produces H_2O vapour and moisturizes the stratosphere. The H_2O vapour is the major source of HOx radicals which can contribute to O_3 loss and controls the hydrogen budget in this region (Rockmann et al., 2004). But in the upper stratosphere, CH_4 reaction is with chlorine or atomic oxygen (McCarthy et al., 2003), the culprits for O_3 destruction, and as a result of this CH_4 moderates the O_3 chemistry in the upper stratosphere by preventing further O_3 destruction.

Several measurement techniques are available to probe the vertical distribution of CH_4 in the atmosphere like satellite-based measurements; aircraft-based gas sampling, gas chromatography, laser spectrometry, etc. The space-based CH_4 studies exploit the infrared absorption bands in solar radiation either in near infrared (NIR) (Yoshida et al., 2011) or terrestrial radiation in thermal infrared (TIR) region of the spectra to retrieve its abundance in the atmosphere (Xiong et al., 2008). Aircraft measurement of CH_4 uses flask sampling or in situ laser absorption spectroscopy techniques (Dyroff et al., 2014). However, limited measurements have been available on the vertical distribution of CH_4 over Indian region in literature (Lal et al., 1994; Patra et al., 2003). Lal et al. (1994) and Patra et al. (2003) studies were based on sample collection by balloon borne cryogenic air sampler in the height 10–35 km at Hyderabad (17° N and 78.6° E). Patra et al. (2003) discussed the influence of quasi biennial oscillation in stratosphere on the CH_4 vertical distribution. The study also pointed out the gradient in tropospheric CH_4 around 10 km and attributed to the vertical transport, but no detailed study is available on this aspect. The present study addresses the seasonal changes in the mixing ratio of the upper troposphere and near surface CH_4 along with the column averaged mixing ratio, over three latitude sectors over Indian region, as observed by aircraft-based, in-situ and satellite based measurements respectively. The observed seasonal features were examined in the light of the airflow pattern/air mass back trajectories, changes in convective activities, vertical winds and boundary layer height (BLH).

2. Data and measurement technique

In addition to the column averaged CH_4 mixing ratio from satellite based SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY), the upper tropospheric CH_4 mixing ratio from Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container (CARIBIC) and the in-situ measured near surface mixing ratio from Cape Rama, Goa the published results on in-situ measurements from Ahmedabad were also used in this study. Table 1 gives the details of the data used.

2.1. Upper tropospheric CH_4 from CARIBIC

The upper tropospheric CH_4 (UCH_4) data over Indian region used in this study is from CARIBIC measurements (http://www.caribic-atmospheric.com). The CARIBIC system (Brenninkmeijer et al., 1999) flown in Lufthansa Airlines Airbus makes measurements of CH_4, O_3, CO, CO_2 etc and aerosol on a monthly basis at an altitude of around 10–12 km. CARIBIC initiated air sampling along Frankfurt to Chennai route in April 2008 (Schuck et al., 2012), and took its last flight to Chennai in March 2012. Air sampling was done during 4 flights in a month, except July and October (only 2 flights during these months). The sample collection is made by using a Triggered Retrospective Air Collector (TRAC) which consisted of a pumping unit and two canister units (Brenninkmeijer et al., 2007). TRAC collected samples at equally spaced intervals of 30–45 min (corresponding to a distance of ~480 km) for 30–90 s (~7–22 km) sampling duration. About 28 samples were collected in one round trip (14 samples onward and 14 samples during return flight). The post flight analysis of CH_4 along with other greenhouse gases CO_2, N_2O and non-methane hydrocarbons were done at Max Planck Institute of Chemistry; Germany using Gas Chromatography-Flame Ionization Detector (GC-FID) system (Schuck et al., 2009) with an average precision of 0.12%. The measurement path (onward and return tracks traversed by the aircraft) from Frankfurt (FRA) to Chennai (MAA) and MAA to FRA for the month of May 2008 are shown in Fig. 1. The rectangle marked in the figure represents the present study region.

2.2. Satellite-based column averaged mixing ratio of CH_4

SCIAMACHY retrieved column averaged mixing ratio of CH_4 (XCH_4) data is used in the present analysis. SCIAMACHY is a passive imaging spectrometer on board ENVIronmental SATellite (ENVISAT) of the European Space Agency (ESA), operated from the 2003 through 2012 period. SCIAMACHY measured reflected, backscattered and transmitted solar radiation in the wavelength range 214–2386 nm at eight spectral channels. The Near Infrared (NIR) window in channel 6 (1631–1671 nm) was used for CH_4 measurement and it suffered degradation after 2005. The measurement continued till 2009 with reduced precision of 4% from 1.7% after November 2005 (Schneising et al., 2012). The data retrieved using Weighted Function Modified-Differential Optical Absorption Spectroscopy (WFM-DOAS) (http://www.iup.uni-bremen.de/sciamachy/NIR-NADIR-WFM-DOAS/products) v2.0.2 algorithm was used in this study (Buchwitz et al., 2006; Schneising e al., 2011). The quality filtered data of XCH_4 with a resolution of $0.5° \times 0.5°$ for the period 2003 through 2009 has been used in the analysis.

2.3. Surface CH_4 measurement at Cape Rama, Goa from CSIRO network

There are several surface observatories for continuous monitoring of gases in the world under the National Oceanic and Atmospheric Administration (NOAA), Advanced Global Atmospheric Gas Experiment (AGAGE), Commonwealth Scientific Industrial Research Organization (CSIRO), University of California Irvine (UCI) with most of their stations located at American and European regions. Out of this, only one CSIRO observatory is located in the Indian region, which is at Cape Rama (15.08° N, 73.83° E), Goa (Fig. 1).

The observatory at Cape Rama, Goa had been operational since February 1993 (Tiwari et al., 2011) as a part of the atmospheric monitoring program, Global Atmospheric Watch (GAW) of the World Meteorological Organization (WMO) for the monitoring of greenhouse gases (CO_2, CH_4, N_2O etc.) and reactive gases (O_3, CO, SO_2, Volatile Organic Compounds (VOCs) etc.) to assess their impact on climate. Sampling location is 60 m above mean sea level and away from major sources/sinks of CH_4. Samples were collected in flasks weekly once and the analysis of the samples were carried out using GC-FID system at CSIRO Marine and Atmospheric Research,