Journal of Environmental Management 289 (2021) 112526

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: http://www.elsevier.com/locate/jenvman

Research article

Biogenic link to the recent increase in atmospheric methane over India

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ARTICLE INFO

Keywords:
Atmospheric methane
GHG Mitigation
Rice farming
Indian agriculture

ABSTRACT

Methane (CH_4) is a prominent Greenhouse Gas (GHG) and its global atmospheric concentration has increased significantly since the year 2007. Anthropogenic CH_4 emissions are projected to be 9390 million metric tonnes by 2020. Here, we present the long–term changes in atmospheric methane over India and suggest possible alternatives to reduce soil emissions from paddy fields. The increase in atmospheric CH_4 concentrations from 2009 to 2020 in India is significant, about 0.0765 ppm/decade. The Indo-Gangetic Plains, Peninsular India and Central India show about 0.075, 0.076 and 0.074 ppm/decade, respectively, in 2009–2020. Seasonal variations in CH_4 emissions depend mostly on agricultural activities and meteorology, and contribution during the agricultural intensive period of Kharif–Rabi (i.e., June–December) is substantial in this regard. The primary reason for agricultural soil emissions is the application of chemical fertilizers to improve crop yield. However, for rice farming, soil amendments involving stable forms of carbon can reduce GHG emissions and improve soil carbon status. High crop production in pot culture experiment resulted in lower potential yield–scaled GHG emissions in rice with biochar supplement. The human impact of global warming induced by agricultural activities could be reduced by using biochar as a natural solution.

1. Introduction

The global climate has experienced a significant warming over the past 100 years (IPCC, 2019; NASA. and GISS, 2018). Studies have shown that CH_4 has 21 times higher heat–trapping potential than that of CO_2, which has resulted in a cumulative gain in radiative forcing of 0.97 Wm^{-2} in sestercentennial (Nisbet et al., 2019; Shindell et al., 2009; Yvon–Durocher et al., 2014). Atmospheric CH_4 has been increasing for the past few decades and this increasing trend has accelerated since 2007 (Zhang et al., 2020). Although the exact sources are uncertain, the increase in CH_4 is due to changes in emissions from wetlands, oceans, soil, fossil fuels, livestock, landfill and termites (Kavitha and Nair, 2017; Potter et al., 2006). This situation demands a significant reduction in global atmospheric methane as a measure to mitigate climate change (Iwata and Okada, 2014).

Methane production is supposed to have diverse fronts, which are identified as biogenic (rice farming, livestock rearing and landfills), anthropogenic (fossil fuels mining and burning), and natural/thermogenic (volcanoes, termites and wetlands). However, increase in atmospheric methane since 2007 have been attributed to biogenic sources (e.g., Zhang et al., 2020). Methane emission occurs due to the actions of methanogenic bacteria on organic matter in an oxygen–limited environment such as puddled soil in paddy fields (Purkait et al., 2005) and piled–up organic wastes dumped in landfill areas (Yan et al., 2009). Rice is an important staple crop in Asia and an important Kharif (JJAS) crop in India (Timsina et al., 2010). Rice cultivation globally accounts for 10–12% of the total CH_4 emissions in the world (IPCC, 2014). Nitrogen (N) fertilizers and anoxic soil in rice fields augment CH_4 emissions (Singh et al., 2018). CH_4 production and oxidation are biologically induced processes influenced directly or indirectly by fertilization (Schimel 2000). The type of N–fertilizer applied also affects the transport of CH_4 from anaerobic soil to the atmosphere (e.g., Le Mer and Roger 2001). The addition of organics like manure or compost to substitute mineral fertilizer also enhances CH_4 emissions (Sánchez–Monedero et al., 2019). Therefore, biochar–based fertilizers that can reduce greenhouse gas (GHG) emissions by modulating soil biogeochemistry and microbial activity are very important in this context (Chojnacka et al., 2020). Xiao et al. (2018) reported a notable decrease in CH_4 emissions from irrigated paddy cultivation with different doses of biochar, owing to the reduction in mineral fertilizer

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https://doi.org/10.1016/j.jenvman.2021.112526
Received 21 December 2020; Received in revised form 26 February 2021; Accepted 30 March 2021
Available online 10 April 2021
0301-4797/

use.

Methane has an atmospheric lifetime short enough to be removed rapidly by oxidation. Besides, photochemical oxidation by hydroxyl radicals (OH–) radicals, the major biochemical pathway for removal of atmospheric CH_4 involves methanotrophic bacteria (Rigby et al., 2017). Etiope et al. (2019) found large uncertainty in the atmospheric CH_4 budget, including sources, sinks and many factors affecting CH_4 emissions (Patra et al., 2011). McGinnis et al. (2016) reported that forest and agricultural lands are equal contributor of atmospheric CH_4 emissions. China and India contribute to a large portion of anthropogenic CH_4 emissions having large seasonal variability (Ganesan et al., 2017). Such emissions are mostly from rice farming, mining activities and other agricultural activities (MoEFCC, 2015). In addition, 10 Mt of CH_4 is released from enteric fermentation, whereas the deep–water rice and excreta of grazing animals contribute an additional 3.27 Mt per annum. Rice farming requires many complex inputs and fertilizers that can further complicate the emission estimates (Patra, 2012). In India, methane concentrations are highest in the Indo–Gangetic plains (IGP) possibly due to rice cultivation (Hayashida et al., 2013; Parashar et al., 1996). However, regional studies are necessary to examine the spatial and temporal variability of atmospheric CH_4 over India.

Space–borne measurements in near or thermal infrared spectral bands are used to complement ground–based studies for atmospheric trace gases (Schaefer et al., 2016). Atmospheric methane measurements are available from AIRS (Atmospheric Infrared Sounder), OCO (Orbiting Carbon Observatory), SCIAMACHY (Scanning Absorption Spectrometer for Atmospheric Cartography), and GOSAT (Greenhouse Gases Observing Satellite) satellite sensors. Although there are long–term ground–based CH_4 measurements, but these are not available for the Indian region. Therefore, we study seasonal, inter–annual, and bi–annual variations of CH_4 over India using the GOSAT data from 2009 to 2020. We also investigate the effect of applying biochar to paddy soils to reduce GHG emissions by substituting some of the mineral N–fertilizers. This study aims to identify the biogenic source of CH_4 and evaluate the role of biochar in reducing GHG emissions from rice fields.

2. Material and methods

GOSAT was launched in January 2009 in a sun–synchronous low Earth orbit. The GOSAT satellite is jointly developed by National Institute for Environmental Studies (NIES), Japan Aerospace Exploration Agency (JAXA), and Ministry of the Environment. GOSAT overpasses at 13:00 (local time) every three days, and the diameter of footprint in nadir is approximately 10 km. As the amplitude of seasonal and annual variability of CH_4 column are small compared to their average atmospheric concentration, the satellite measurements should provide a demanding precision of 2% (Zhou et al., 2016). Since GOSAT has a high pixel resolution and precision (10×10 km^2 and 0.6%), the measurements are suitable for analysing temporal and spatial changes in atmospheric CH_4 (Turner et al., 2015). From the TANSO–FTS (Thermal and Near Infrared Sensor for carbon Observation–Fourier Transform Spectrometer) instrument onboard the GOSAT, CH_4 columns have been continuously available since May 2009 (Kuze et al., 2020). To identify the cloudy pixel, it has a dedicated Cloud and Aerosol imager (CAI). The TANSO–FTS instrument has 4 spectral bands with a high spectral resolution of 0.2 cm^{-1}, three of which operate in the SWIR at around 0.76, 1.6 and 2.0 μm providing sensitivity to the near–surface absorbers, with the fourth channel operating in the thermal infrared between 5.5 and 14.3 μm providing mid–tropospheric sensitivity. The SWIR retrieval algorithm consists of three steps: data screening suitable for the retrieval analyses, optimal estimation of gaseous column abundances and finally checking the quality of retrieval results. During the retrieval process to avoid cloud contamination, the scene with greater than one cloudy pixel is excluded. The cloudy pixels are identified from the atmospheric images of CAI. Due to this strict screening, a relatively smaller frequency of XCH_4 measurements is available during the monsoon (JJAS) period over south Asia (Chandra et al., 2017). The SWIR retrieval algorithm is explained in detail by Yoshida et al. (2011).

Several algorithms have been developed to retrieve GOSAT XCH_4, such as University of Leicester full physics retrieval algorithm OCFP and proxy version OCPR (Schepers et al., 2012), Netherlands Institute for Space Research/Karlsruhe Institute of Technology (SRON/KIT) full physics retrieval algorithm SRFP and proxy version SRPR (Butz et al., 2011), and the NIES algorithm (Yoshida et al., 2013). All GOSAT XCH_4 retrieval algorithms have already been validated using the TCCON (Total Column Carbon Observation Network) observations (e.g., Yoshida et al., 2013; Parker et al., 2011; Butz et al., 2011; Cressot et al., 2014), and the validations suggest that the GOSAT data can be used for studies on seasonal variability and long–term trends (Kivimäki et al., 2019). The GOSAT data were also used to study the variability in atmospheric CH_4 over India (Chandra et al., 2017; Ganesan et al, 2015, 2017; Prasad et al., 2014). Here, we have used the NIES full physics SWIR Level 3 retrieval algorithm version 02.80 of XCH_4 over the Indian region. The FTS SWIR Level 3 data products are generated by interpolating, extrapolating and smoothing the FTS SWIR Level 2 column–averaged mixing ratios of CH_4 on a monthly basis. The values are gridded to 2.5° cells and the XCH_4 data have a mean bias of −1.9 ppb (Morino et al., 2019).

The rice area and rice production data from 2009 to 2020 are taken from DES (Directorate of Economics and Statistics) India. Livestock data (Sonavale et al., 2020) for the period 2009–2020 are taken from the livestock reports (2012 and 2019) of Department of Animal Husbandry, Dairying and Fisheries, India. The wetlands data (Bassi et al., 2014) are taken from National Wetland Inventory and Assessment (NWIA) of the Ministry of Environment and Forest (MoEF). Coal production data for the period 2009–2020 are collected from the Ministry of Coal, Government of India. The fertilizer and manure consumption data from 2009 to 2020 for India are taken from the FAOSTAT (Food and Agriculture Organization Corporate Statistical Database) data portal. We have also considered the cloud corrected fire count data from MODIS (MODerate Resolution Imaging Spectrometer) on Terra and Aqua in 2009–2020. These datasets have a spatial resolution of $0.5^\circ \times 0.5^\circ$ suitable to study the relationship between methane emissions and biomass burning. Here, we have used the MOD11C3 product of the Land surface temperature (LST) to study the connection between CH_4 emissions and change in the ground temperature (Javadinejad et al., 2019). Soil moisture data over India from 2009 to 2018 are obtained by averaging the 'Surface Soil Moisture' data products, from the Bhuvan portal of ISRO (Indian Space Research Organisation). Soil organic carbon (SOC) data used in our study are obtained from SoilGrids, which is a global soil information system containing spatial predictions for several soil properties (clay, silt and sand content, pH index, cation-exchange capacity), at seven standard depths: 0, 5, 15, 30, 60, 100 and 200 cm (Hengl et al., 2017). The wind data are taken from the ERA–5, and the data have a spatial resolution of 0.25° x 0.25° (Hersbach et al., 2020).

A controlled environment greenhouse study was conducted in a completely randomized experimental design using undisturbed soil, with different biochar and compost doses as a substitute to mineral fertilizers in integrated nutrient management of paddy. The undisturbed soil is assumed to have a stable microbial community structure contributing to the soil emissions (Goberna et al., 2005). The treatments comprise a different combination of biochar/compost with mineral fertilizers in six replications, as shown in Table 1. The chemical analysis of experimental soil and environmental conditions are explained in Table S1. The experimental soil belongs to the Vertisol order of U.S. soil Taxonomy and filled in 1300 cm^3 pots, which were planted with two (21 days old) rice plants per pot after taking out from nursery. The crop management and yield estimation were done during crop growth and maturity, respectively (Nayak et al., 2020). The nutrients N, P and K were supplied at a rate of 125, 62.5, and 45 kg ha^{-1} of N, P, and K, respectively, as soil test based (STB) recommendation in the form of Urea, Single superphosphate (SSP), and Muriate of potash (MOP) as per

Table 1
Potential yield scaled GHG of nutrient management treatments for cultivation of paddy.

T. No.	Nutrient management treatments	Abbreviation	Urea	Soil Organic Carbon	Potential emission	Grain yield	Yield scaled GHG
			(g/pot)	(%)	g CO_2–C[a]	g pot^{-1}	gCO_2–C g^{-1} (10^{-3})
T1	Soil–test based N (urea) fertilization	CF_{STB}	1.72	0.28±0.05	0.344	60.38±1.08	5.62±1.08
T2	Low–dose biochar INM¶ (75% N(urea) + biochar @ 3 Mg ha^{-1})	CF_{STB75} + BC_{25}	1.29	0.50±0.04	0.258	89.3±1.33	2.81±1.33
T3	Low–dose compost INM (75% N(urea) + compost @ 3 Mg ha^{-1})	CF_{STB75} + CO_{25}	1.29	0.48±0.05	0.258	65.92±1.50	3.92±1.50
T4	High–dose biochar INM (50% N(urea) + biochar @ 6 Mg ha^{-1})	CF_{STB50} + BC_{50}	0.86	0.44±0.05	0.172	66.56±1.60	2.59±1.60
T5	High–dose compost INM (50% N(urea) + compost @ 6 Mg ha^{-1})	CF_{STB50} + CO_{50}	0.86	0.50±0.01	0.172	46.7±1.05	3.63±1.05

INM¶ means Integrated nutrient management, g CO_2–C[a] means gram CO_2–C calculated using IPCC emission coefficient for Urea (Kim et al., 2016). Results are expressed as mean±standard deviation.

the treatment details. The mineral fertilizers were applied as concentrated solution evenly in pots, the day before planting, while organic fertilizers (biochar/compost) were mixed thoroughly in the topsoil (5 cm). All mineral fertilizers were applied basal as of their full dose except mineral N in 2 splits with 50% as basal and 50% at the panicle development stage. The modified IPCC GHG emission coefficients for mineral fertilizers in soils were considered to calculate the potential GHG emission per gram of rice grain produced in each treatment and presented as potential yield–scaled GHG emission (Gupta et al., 2009; Kim et al., 2016; Stephen et al., 2019).

3. Results and discussion

3.1. Inter–annual variability

The annual atmospheric methane concentration over India from 2009 to 2020 is shown in Fig. 1. The lowest methane concentration is observed in the Hilly region (ice–covered and barren regions of Kashmir). However, the concentration increases from Hilly to IGP regions of India. This is due to the intensive rice cultivation during the monsoon in this region. Moreover, the highest concentration of methane is observed in Central India and it decreases towards the Peninsular region. In addition, the annual methane concentration shows a significant increasing trend (0.0765 ppm/dec; $p < 0.05$) from 2009 to 2020 over India (Fig. 2). Furthermore, IGP, Peninsular and Central India show

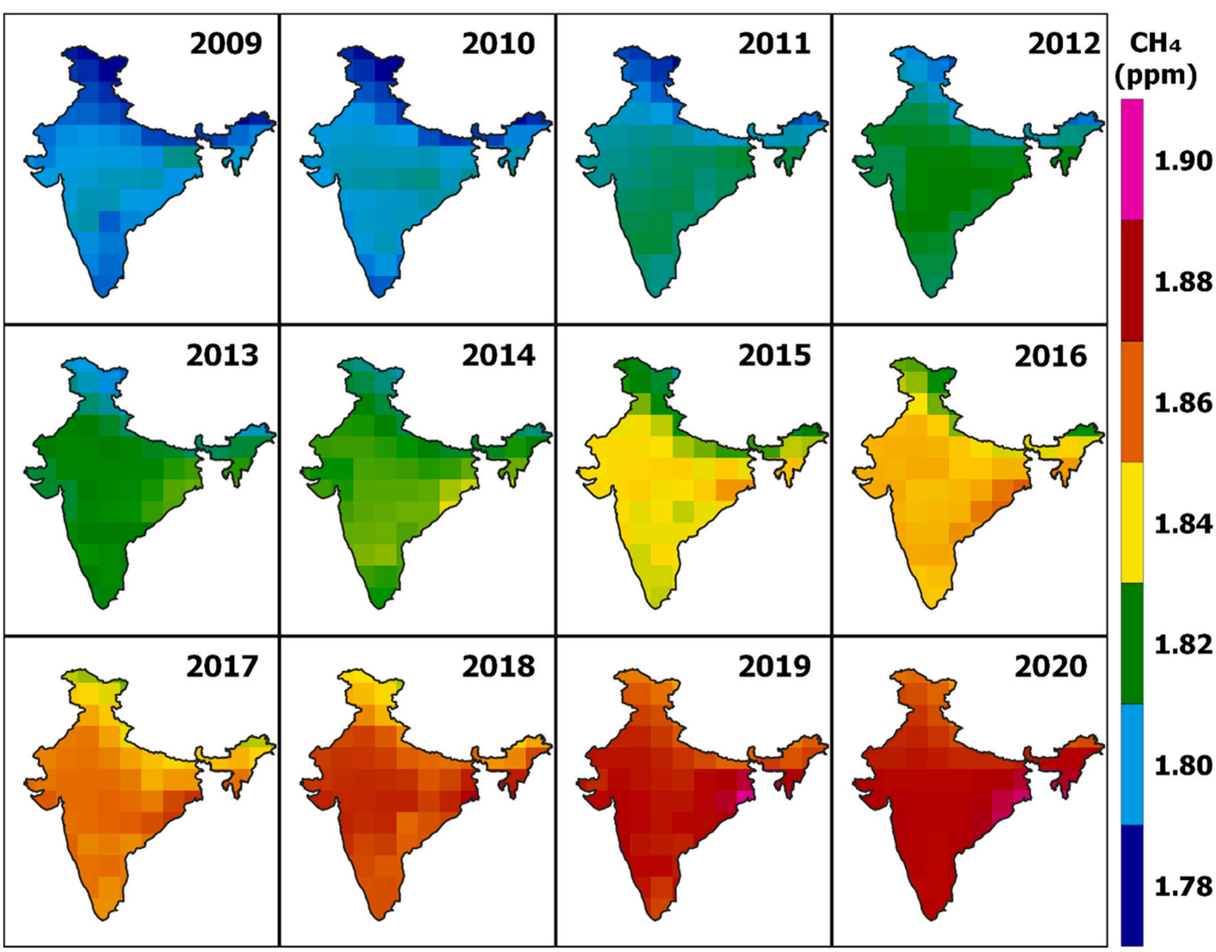

Fig. 1. Increasing Methane. Atmospheric methane over India from 2009 to 2020 from the GOSAT satellite measurements.

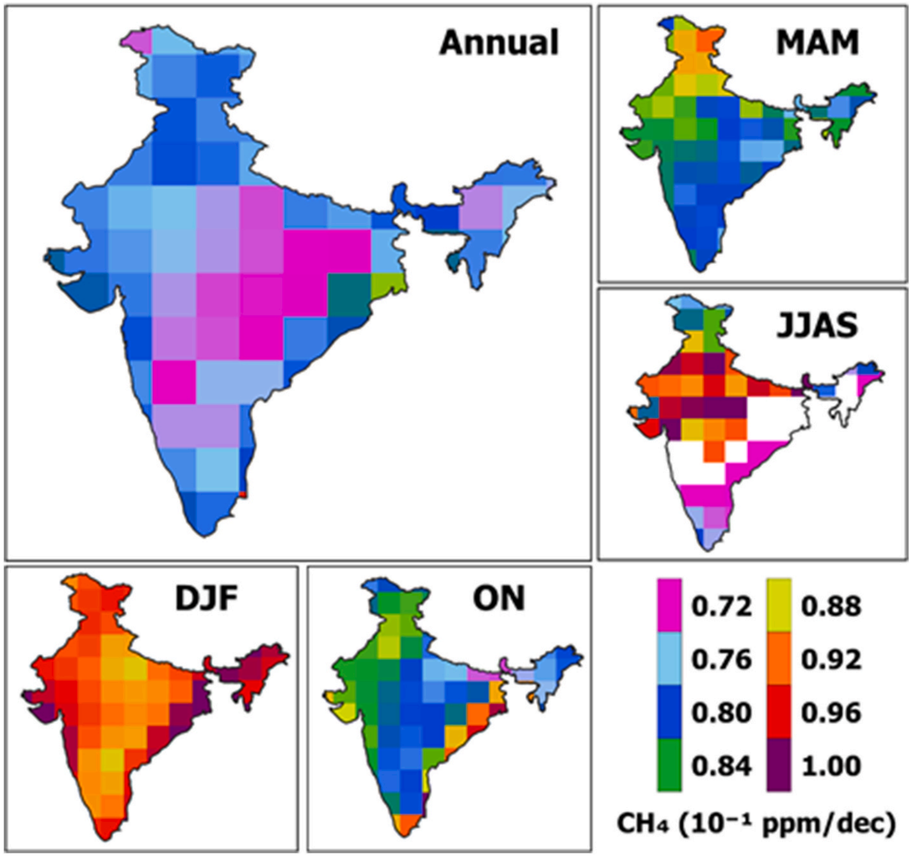

Fig. 2. Trends in Atmospheric Methane. The GOSAT CH_4 trends estimated from measurements in different seasons [i.e. winter (DJF), pre–monsoon (MAM), monsoon (JJAS), post–monsoon (ON)] and annual average over India from year 2009–2020 (dec. = decade). All the trends are statistically significant at 95% CI.

a significant increase of about 0.075, 0.076 and 0.074 ppm/dec, respectively, in their annual methane concentration from 2009 to 2020. The major drivers for the increase in methane over India are arable land under rice cultivation (Parashar et al., 1996), livestock population (i.e., there is an increase of about 14 million in livestock population from 2012 to 2019 in the Peninsular region), chemical fertilizer consumption, wetland emission, and emissions from mining activities (Kavitha and Nair, 2016). However, the different regions of India, as presented in Fig. S1, show a statistically significant ($p < 0.001$) increasing trend over the period. The uncertainty in trend values in different regions indicates the influence of prominent local driver of methane. For instance, North–Eastern India (NEI) shows the highest (0.079 ppm/dec), but Central India shows the smallest (0.074 ppm/dec) rate of increase from 2009 to 2020. Although both regions show a significant increase in methane concentration, the difference in magnitude can be attributed to meteorology, tropical evergreen forests, and high SOC in NEI, whereas the preference for coarse–grain crop cultivation in the Kharif season in Central India. This is also supported by the land use land cover (LULC) map of India that shows dominance of forest cover in NEI, but croplands in Central India (Fig. S2, Left panel).

Paddy cultivation is one of the key contributors to methane emissions, as reported by several studies (Chandra et al., 2017; Zhang et al., 2020). This is also shown by the correlation of increasing arable land area under rice cultivation and increase in methane emission in North–West India (NWI, $r = 0.78$), IGP ($r = 0.72$), and Central India ($r = 0.69$) (Table S2a). Other agricultural activities, such as livestock rearing, manure and compost production, along with mineral nitrogen fertilization in saturated soils also contribute to methane emissions (Kavitha and Nair, 2017). The emission of methane is strongly influenced by soil bio–geochemistry, which depends on soil pH, soil temperature and SOC. The increase in the organic carbon in croplands in the past decades (Nayak et al., 2020) indicates the increased use of organic fertilizers and soil conditioners. The production of organic manures ($r = 0.98$) and (Table S2b) the use of chemical fertilizers ($r = 0.61$) in paddy increases CH_4 emissions (Galic et al., 2020). The direct stimulation impact of N–fertilization on CH_4 has also been observed (Dan et al., 2001). When analysing the impact of application of chemical N–fertilizer on CH_4 emissions from rice fields, it is reasonable to conclude that N–fertilizer usage will increase the biomass of rice, including the output of CH_4 (e.g., Schimel, 2000). Our results show a significant increase in the livestock population in some parts of Lower IGP (West–Bengal, Bihar and Jharkhand Fig. S3), Central India (Maharashtra, Madhya Pradesh and Chhattisgarh), and Peninsular India (Karnataka, Kerala and Tamil Nadu) (Fig. 3, Lower panel). There is a 4.7% increase in the total livestock population from 2012 to 2019 in India, which greatly contributes to the increasing trends in CH_4 during the period. In addition, the increase in coal production (0.23%) across years (2009–2019) in the eastern part of India (i.e., Jharkhand) while crude oil mining in the coastal regions might be a key factor for CH_4 emissions. The North–Western region has the largest area under wetlands (e.g., Gujarat has the largest area under wetlands as reported by Bassi et al., 2014), which contribute to the CH_4 emission in that region.

The effect of meteorological factors such as wind speed and direction (Fig. S4) can also be observed in the CH_4 concentration. There is a contribution of the wind to the higher concentration of methane observed over the Peninsular region during the October–November (ON) season, as the wind blows from the northern and Central India towards Peninsular India in this season (e.g., Kavitha and Nair, 2017). The effect of intense agricultural activities in the Indus river valley (i.e., at foothills of Himalaya) in the case of northern Hilly regions can be found in the analyses. The agricultural activities are seasonal and thus, a distinct seasonal variability is evident in the CH_4 concentrations. The

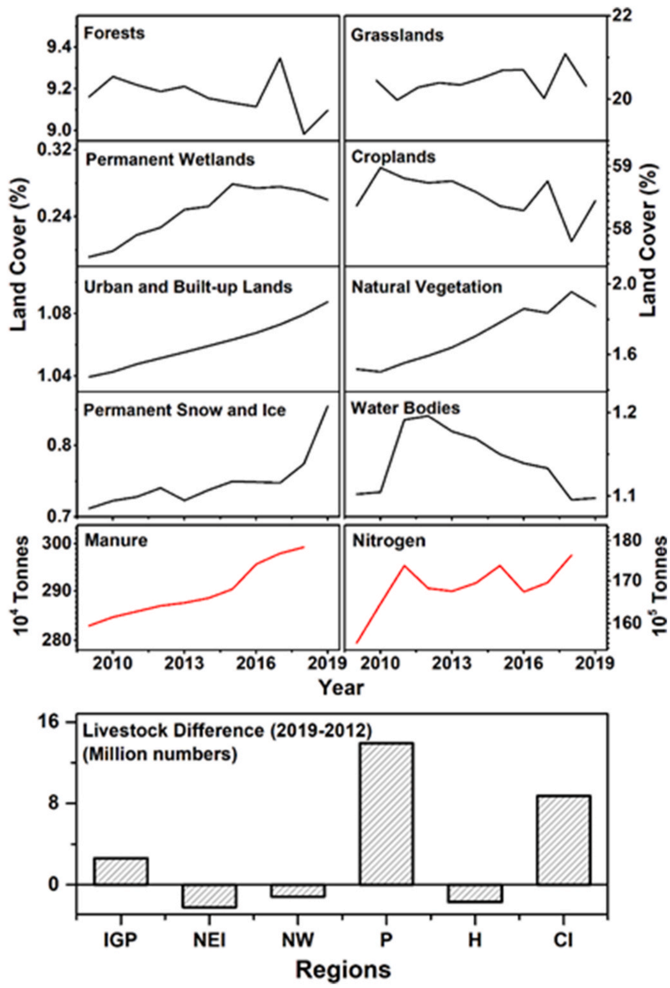

Fig. 3. The LULC Changes. (Upper panel) The inter–annual variation of different type of LULC (Land use Land cover) pattern from 2009 to 2019 in India. (Middle panel) Inter–annual variation of manure production and nitrogen fertilizer consumption from 2009 to 2018 in India. (Lower panel) Livestock difference between 2019 and 2012 in different regions (as illustrated in Fig. S1) of India.

burning of coal and wood also produces CH_4 and henceforth, the forest fires and energy production through thermal power plants contribute to the high CH_4 concentrations in the lower IGP (Miller et al., 2019). The north–eastern Hilly regions show high concentrations of CH_4, which can be due to the increased incidences of forest fires (Chakraborty et al., 2014), as illustrated in Fig. 4.

3.2. Seasonal variability

Seasonal variability in CH_4 emissions depends mostly on agricultural activities and meteorology. We analysed the distribution of atmospheric CH_4 over India in all four seasons (i.e., MAM or March–April– May, JJAS, ON, DJF or December–January–February) in the period 2009–2020 (Fig. 5). The Land use and Land cover pattern in India is also analysed (LULC, Fig. S2 Left panel), as the agriculture and land use pattern are substantial contributors of CH_4 emissions (Harper et al., 2018). Higher CH_4 concentration lie over croplands of IGP and Central India croplands, due to intensive rice cultivation during Kharif (monsoon) season. Wetlands and urban built–up areas are the potential emitters of CH_4 over the North–Western region, whereas forests and natural vegetation contribute to emissions over NE India, as depicted in the LULC analyses (Kuttippurath, 2021a,b). Urban built–up, permanent wetlands and natural vegetation show an increasing trend (Fig. 3, Upper panel) from 2009 to 2019 and contribute to the increasing trends of methane in the last decade. The Hilly region shows the smallest CH_4 concentrations compared to other regions of India because of the snow and ice cover round the year. However, analyses show that increase in vegetation cover is an important factor for the increase in CH_4 concentration over India as the decomposition of plant litter and CH_4 emission are closely related (Hayashida et al., 2013).

The burning of biomass also triggers methane emissions (Grutzen and Andreae, 1990). Burning of crop stubbles (Fig. 4) in post Kharif season is practiced (particularly in IGP) to save time for Rabi sowing (Kuttippurath et al., 2020). In our analyses, the highest concentrations of methane are found in ON, which indicates the combined effects of biomass burning in the post–harvest season of rice in northern India (particularly IGP), the cultivation of rice in the south–west monsoon season in the east coastal regions of Peninsular India, and the year–round cultivation of rice in the lower IGP and NEI. Apart from that, the effect of monsoon winds blowing from Bay of Bengal to the landmass also brings the methane from the ocean (e.g., Kavitha and Nair, 2016). The lowest concentration of CH_4 is found in MAM, which is an agricultural non–intensive season, and most croplands remain fallow during this period. In most parts of IGP, and North–Western India, farmers do not perform any field activities in this period. However, the regions around the east coast and northeast India (e.g., Orissa, West Bengal and Assam) follow sequential summer rice cropping there. These can be the reasons for the high methane concentration in those regions.

Rice is a staple cereal food in India and cultivated mostly in JJAS (Kharif) season. Almost 84% of arable land in India undergoes rice farming during this period (State of Indian Agriculture, 2017) The incubation of Louisiana rice soil adjusted with rice straw under anaerobic conditions by Wang et al. (1992) also showed a stimulating effect of urea on the increase in CH_4. The Kharif rice might be the primary cause of high CH_4 concentration in IGP and parts of Madhya Pradesh, Maharashtra, Andhra Pradesh during JJAS season. The regions affected by the south–west monsoon in India show a higher rate of increase (0.09–0.1 ppm/dec) in JJAS, which indicates the influence of meteorology, soil moisture and rice cropping (Hayashida et al., 2013) in that period. However, a relatively lower rate of 0.07–0.08 ppm/dec can be observed in Peninsular India (e.g., Tamil Nadu), which mostly receives rain during the northeast monsoon (ON) season.

Wheat or legumes/oilseeds are mostly cultivated in Rabi season (DJF), and are normally irrigated during critical crop growth stages. The soil remains aerobic during most of the season, unlike paddy, which has anaerobic soil favouring soil methanogenesis. The aerobic soil mostly favours the oxidation of native SOC and thus, CH_4 production is very unlikely in such conditions as shown in DJF season in most regions of India (Muñoz–Rojas et al., 2013). However, the rate of increase is highest (approx. 0.8–1%) in ON–DJF across India, which is due to the opening of soil furrow for sowing and intercultural operations of Rabi crops and that would release the trapped CH_4 in anaerobic soil pockets. Yet, other factors like coal mining (i.e., Jharkhand) can add to the high concentrations of CH_4 along with summer rice cultivation in the eastern coastal plains. However, a bi–annual analysis can separate the role of agricultural sources from non–agricultural sources of methane emissions.

3.3. Bi–annual variation

The bi–annual CH_4 analyses (Fig. 6) show that the agricultural intensive period of Kharif – Rabi (i.e., June–December) is a significant contributor to CH_4. Methane produced by methanogens in the soil is strongly influenced by the amount and type of fertilizer and/or amendment applied during the cropping season (Yuan et al., 2018). The mineral fertilizers used in the submerged soils might be a readily available nitrogen source to the microbes to convert soil carbon to CH_4 (Bargaz et al., 2018). Additionally, at the microbial community level, the application of N to soil would not only facilitate the growth and

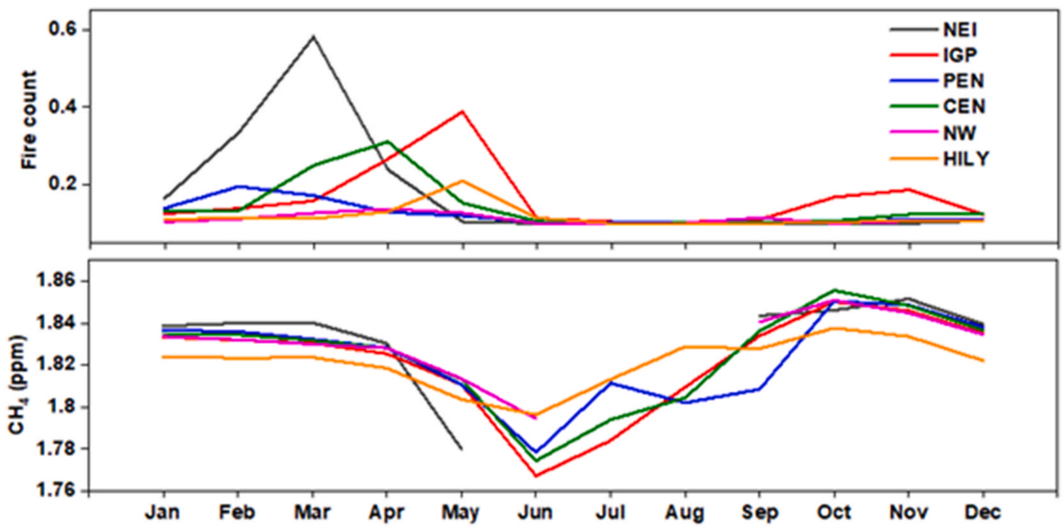

Fig. 4. Fire events. The monthly distribution of fire count (upper panel) and CH_4 (lower panel) at different regions of India [IGP, Peninsular, North West, Central India, NEI and Hilly, as illustrated in Fig. S1]. The data are averaged from year 2009–2020.

production of microbes, but would also increase the growth and operation of methanogens (Schimel (2000). However, organic fertilizers and farmyard manures also contribute significantly to CH_4 emissions (Skinner et al., 2019). We analysed the amount of organic manure produced and mineral fertilizers consumed during the period 2009–2020. The increase in organic manure production and chemical fertilizers consumption (Fig. 3, Middle panel) clearly indicate the possible increase in CH_4 emissions.

Methanogenesis in soil also depends on the soil moisture and native soil organic matter (e.g., Yuan et al., 2019). We analysed the average soil moisture for agricultural intensive (July–December) and non–intensive (January–June) period from 2009 to 2020, and found that the agricultural intensive period has a comparatively higher (0.12–0.19 m^3/m^3) soil moisture over the croplands. The water–filled pore spaces in soil may favour the soil methanogenesis and thus correlate positively with moisture in the soils (Smith et al., 2003). However, low moisture soils in the agricultural non–intensive periods have more air–filled pores spaces that act as a sink for CH_4 (Fiedler et al., 2005). The low moisture in the soils during agricultural non–intensive period can also be attributed to higher LST during the period, as shown in Fig. 6.

The soil organic matter (SOM) is an another important driver for soil methanogenesis (Corbett et al., 2015). However, the CH_4 emissions have been reported to vary with the amount and quality of carbon input in soil (Bernal et al., 2017). We analysed the SOC stock and found that high emissions from the croplands might have resulted in low to moderate (15–45 t/ha) level of organic carbon (Fig. S2, Right panel). This suggests that the conventional ways to sequester carbon in soil need policy level interventions, as they contribute to enhanced CH_4 emissions from croplands. However, the modelling study by Sreenivas et al. (2016) predicts a possible increase in the SOC in croplands, which seems insignificant with the increasing atmospheric CH_4 and subsequent global warming. The increase in soil temperature with global warming may further reduce the SOC stock through enhanced decomposition of native organic matter (Brevik, 2013; Kirschbaum, 2000).

Mineral fertilizers play a significant role in the fast decomposition of SOM, and thus efforts are needed to reduce mineral fertilizer use (Zhao et al., 2020). There are studies reporting the effects of fertilizer N–application on potential CH_4 emissions from the rice soil, and therefore, more reasonable management practices aiming at efficient use of N inputs are needed, in particular. However, the efforts to reduce the mineral fertilizer use in croplands should be sustainable and must have little impact on food production to ensure food security. Fertilizer alternatives to conventional organic fertilizers such as compost and vermicompost are needed as they also largely contribute to CH_4 emissions during pre– and post–application to soils (Singh Rajeev Pratap, 2012). The organic soil amendments have been reported to modify the microbial community structure/physiological functions and thus, influence soil emissions (e.g., Gorovtsov et al., 2020; Mackelprang et al., 2018). However, comparatively stable carbon alternatives of organic fertilizers, e.g., biochar–based, are reported as an effective tool to reduce CH_4 emissions and improve the soil carbon status (Huang et al., 2019). The major apprehension in the case of biochar fertilizers might be the reduction in grain yield in the case of paddy (e.g., Ali et al., 2020; El–Naggar et al., 2019).

3.4. Comparative evaluation of fertilizations option in paddy to reduce GHG emissions

The pilot scale–controlled environment study conducted to compare the efficacy of conventional compost and biochar fertilizers are presented in Table 1. It shows that biochar–based fertilizers can reduce the mineral fertilizer use and improve grain yield of paddy with improved SOC at harvest. Therefore, it can out–perform the conventional compost in integrated nutrient management strategies for paddy. Our experimental results revealed that reducing the mineral nitrogen fertilizer doses by 25–50% using biochar and compost can reduce the potential yield–scaled GHG emissions by 50–53% and 30–35%, respectively. The reduction in overall potential yield–scaled GHG emissions using biochar as compared to the conventional compost as an alternative to chemical fertilizer in integrated nutrient management of rice can cut emissions as compared to sole mineral fertilizer. However, the gain in rice grain yield varied from 10 to 47% with a 25–50% reduction in chemical–N use, respectively (Fig. S5). The pot experiment results are used as base results in the past to extrapolate the effect of different adjuvants to soil on rice grain yield and growth of paddy crop through a subsequent field trial (eg., Nayak et al., 2019; Yin et al., 2016). Paddy CH_4 emissions have been found to have decreased dramatically as a result of biochar alteration, which, surprisingly, did not result from the inhibition of methanogen (Feng et al., 2012). In contrast, the application of compost or manure such as farmyard manure (FYM) enhances the emission of methane by introducing organic carbon and N needed for microbial activities and acting as a supply of electrons, as reported by Pathak et al. (2003). Similar findings are reported while substituting 50% of inorganic N with FYM, which raised the soil emission by 172% relative to the application of complete N using urea fertilizer (Pathak et al., 2005). Black soils emit an average of 1100 kg/ha of CH_4 from rice cultivation

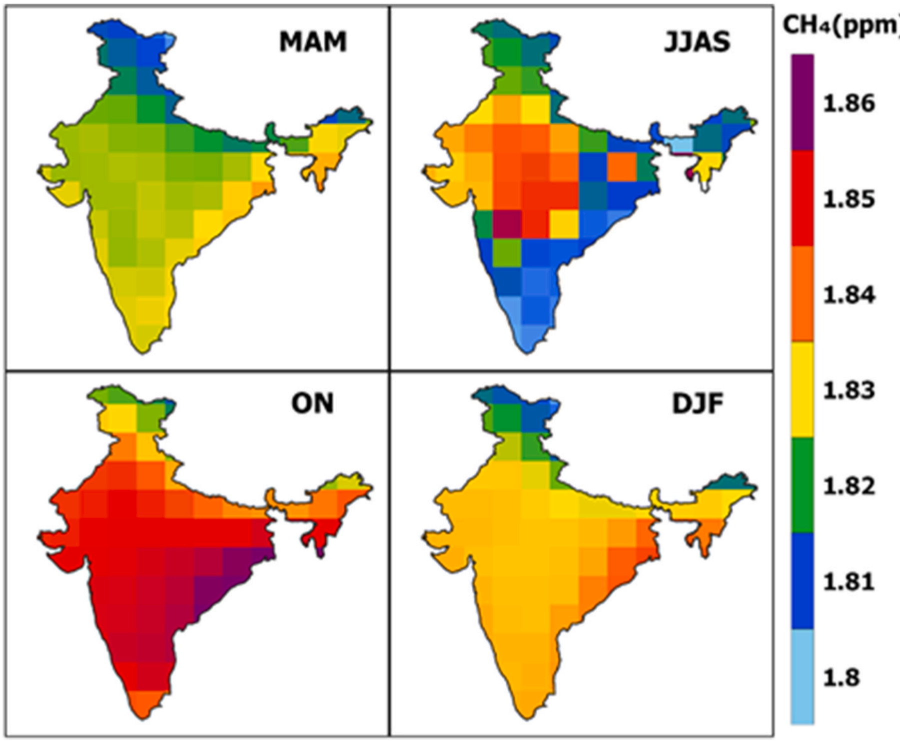

Fig. 5. Seasonal Changes in Methane. Seasonal distribution of atmospheric methane concentration from 2009 to 2020 over Indian region: MAM (Pre–monsoon), JJAS (Monsoon), ON (Post–monsoon) and DJF (Winter).

(Bhatia et al., 2010), which can be due to high soil pH (Wang et al., 1992). When compared with compost as a tool to reduce the GHG of paddy, better efficacy of biochar can be a useful indicator for policy intervention aimed at reducing the carbon footprints of agriculture.

4. Conclusion

The seasonal and inter–annual variability and trends in atmospheric methane during the period 2009–2020 are assessed using the GOSAT datasets. Agriculture (a major biogenic source), in its conventional form, contributes significantly to the atmospheric emission of CH_4. The highest CH_4 concentration is observed over northern, Peninsular and Central India during post–monsoon (ON) season due to receding soil moisture from highly cultivated farmlands and release of CH_4 from anaerobic soil pockets. In contrast, the winter season (DJF) shows a reduction in CH_4 concentration over India because of Rabi farming, which is performed with aerobic soil and scavenging of CH_4 due to photochemical and biochemical reactions. CH_4 concentrations in atmosphere particularly in winter, could have detrimental effects on the respiratory health of humans, ecosystems and climate; suggesting the importance of the analyses presented in this study. However, our analyses infer that low LST and high soil moisture during the agricultural intensive period (Kharif–Rabi) favours CH_4 emission. The total fertilizer consumption is positively correlated with CH_4 formation in the soil; implying that the methanogenesis in the soils is strongly influenced by the changes in agricultural practices i.e., type of fertilizers/amendments used. The IGP, Peninsular, Central India and the whole India show a significant increase of about 0.075, 0.076, 0.074 and 0.0765 ppm/dec, respectively, in their annual CH_4 concentrations from 2009 to 2020. Therefore, efforts to reduce N–fertilizer use in agricultural intensive seasons are required, especially in the IGP, Peninsular, and Central India. Regulations on fertilizer application during cropping seasons in arable lands are thus suggested to cut agricultural emissions. Our study further emphasizes that the N–fertilizers being applied on soil test–based recommendations in rice farming should also be further optimised using stable carbon soil amendments to reduce soil emissions. However, the reduction in yield–scaled GHG emissions using biochar as soil amendment can be well–extrapolated to a larger scale through a field study for better policy decisions. Nevertheless, our study shows a connection between atmospheric CH_4 emission and the use of organics for soil health; bolstering the theoretical understanding of the mechanisms involved in soil methanogenesis. The findings lay down a foundation for a mechanistic study comparing efficacy of different stable carbon alternatives in reducing the potential soil emissions.

Data availability

The data used in this study are publicly available. The GOSAT satellite data are available on https://data2.gosat.nies.go.jp/GosatDataArchiveService/usr/download/DownloadPage/view. The MODIS fire data are on https://neo.sci.gsfc.nasa.gov/view.php?datasetId=MOD14A1_M_FIRE. The ERA–interrim data are available on: https://www.ecmwf.int/en/forecasts/datasets/reanalysis–datasets/era–interim. The fertilizer consumption data are taken from http://www.fao.org/faostat/en/#data. The MODIS Land surface temperature (LST) data are available at https://neo.sci.gsfc.nasa.gov/view.php?datasetId=MOD_LSTD_M&year=2020. The Soil moisture data are available at https://bhuvanapp3.nrsc.gov.in/data. The soil organic carbon data are available on https://soilgrids.org/.

Credit statements

The research was conceived and designed by JK. The first draft was made by AS, KA and JK. All authors contributed to the discussion and a subsequent final draft was made. The data analysis was done by AS. The pot-experiments and analyses are conducted by KA, which were supervised by SD and GC. The figures were made by AS, JK, SR, and KA. The research supervision and project management were done by JK and NM.

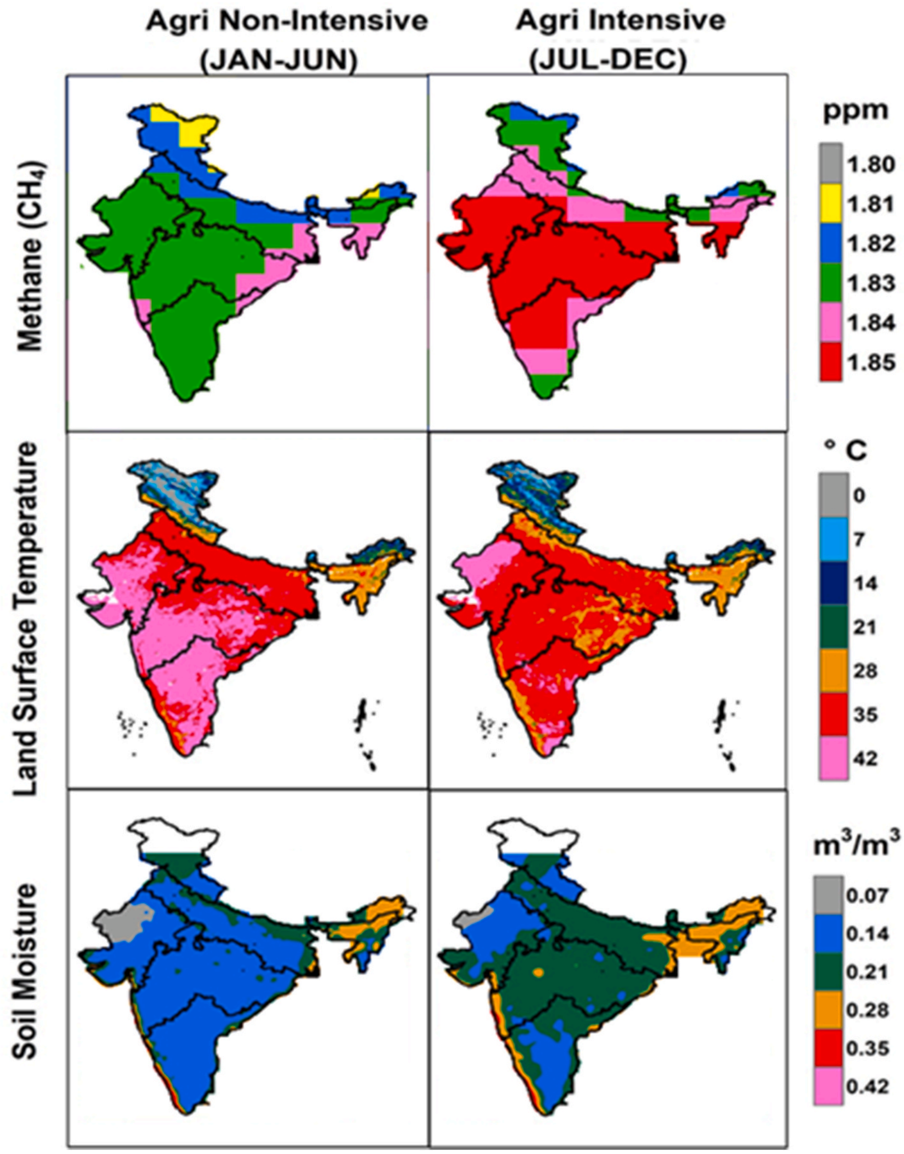

Fig. 6. The factors controlling Methane emissions. Bi–annual distribution of atmospheric methane, LST (Land Surface Temperature) and Soil moisture in Indian from 2009 to 2020. Left: Agriculture non–intensive period, January–June (Dry period). Right: Agriculture intensive period, July — December (Wet period).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We thank the Head, CORAL and AgFE; and the Director, Indian Institute of Technology Kharagpur, Ministry of Education (MoE); Naval Research Board (OEP), Défense Research Development Organization; Indian National Centre for Ocean Information Services (INCOIS) Hyderabad (O–MASCOT project) and Ministry of Earth Science (MoES) for facilitating and funding the study. AS, KA and SR thank MoE, Government of India for their fellowship. We also thank all the data managers and the scientists who made available those data for this study. We thank IMD (India Meteorological Department/MoES) for precipitation and temperature data. The MODIS datasets were acquired from the Level–3 and Atmosphere Archive and Distribution System (LAADS) Distributed Active Archive Center (DAAC), located in the Goddard Space Flight Center in Greenbelt, Maryland (https://ladsweb.nascom.nasa.gov/). We also acknowledge Directorate of Economics and Statistics, Department of Agriculture and Cooperation, India; Department of Animal Husbandry, Dairying & Fisheries, India; Ministry of Petroleum and Natural Gas, India for the agricultural, livestock and gas and oil statistical data. We would like to thank the entire GOSAT team for their dedicated and continuous efforts in producing the data for all these years. We acknowledge the assistance provided by the International Crops Research Institute for Semiarid Tropics (ICRISAT) and ICRISAT Development Centre (IDC) for providing all required experimental facilities.

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