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Sulphur-coated urea reduces greenhouse gas intensity and enhances soil quality in rice cultivation

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ABSTRACT

The Indo-Gangetic Plain (IGP) faces significant challenges related to greenhouse gas (GHG) emissions and declining soil health due to intensive rice-based cultivation systems. This study evaluated the efficacy of enhanced efficiency fertilizers (EEF), including slow-release fertilizer (Sulphur-coated urea, SCU), and nitrification inhibitors (Neem-coated urea, NCU; Karanj-coated urea, KCU) in reducing GHG intensity and improving soil biological activity in rice systems in the IGP. Field experiments conducted over two years assessed yield parameters, GHG emissions, and indicators of soil microbial biomass, nutrient content, and enzymatic activity. NCU reduced CH_4 emissions by 11 % and N_2O emissions by 16.5 % relative to prilled urea (PU), while SCU and KCU also demonstrated notable emission reductions. Sulphur coated urea demonstrated the lowest greenhouse gas intensity (GHGi) (0.128 kg CO_2-eq kg^{-1} grain yield), followed by NCU and KCU. All EEFs significantly improved rice grain yield compared to PU, with SCU and KCU recording the highest mean yields (~5600 and ~5560 kg ha^{-1}, respectively) versus 5010 kg ha^{-1} under PU. Additionally, EEFs improved microbial biomass carbon and nitrogen, dehydrogenase activity, and reduced nitrate reductase and urease activity compared to conventional prilled urea (PU), with KCU and SCU showing the greatest improvements and highest net returns. Among the EEFs, SCU consistently achieved the highest yield, lowest GHGi, and overall improvements in soil health, making it a promising alternative for sustainable rice production. Projections indicate that while application of NCU in the IGP region during rice cultivation could reduce the GHGi by 12.2 % while adopting SCU may achieve a 25.8 % reduction, supporting India's commitment to the Paris Climate Agreement and promoting sustainable agricultural practices in the IGP.

1. Introduction

The Indo-Gangetic Plain (IGP) region is dominated by extensive Rice-Wheat cropping system (RWCS) and spans around 13.5 to 14 million hectares [1], with 10 million hectares in India alone [2]. However, the IGP faces significant challenges pertaining to high greenhouse gas (GHG) emissions and declining soil health [3–5]. Despite occupying a large part of the IGP, the sustainability of the RWCS has become a critical issue due to the fact that yields of both rice and wheat have not further increased over last few decades [4]. Factors such as intensive tillage [6], crop residue burning [7], indiscriminate use of nitrogenous fertilizers [8], and extensive rice-wheat rotational farming have exacerbated the deterioration of soil and air quality [9]. These practices have led to a decline in the soil organic carbon pool, soil biodiversity and increased soil compaction [10]. They have also resulted in significant emissions of potent GHGs such as methane (CH_4) and nitrous oxide (N_2O) [8,11]. Emissions of CH_4 from rice paddies are estimated between 24 and 40 Tg of CH_4 annually, representing roughly 8 % of global anthropogenic CH_4 emissions [8,12].

While CH_4 is the end product of decomposition of organic matter,

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N_2O on the other hand is primarily produced during nitrification and denitrification processes as intermediate product [13,14]. Under aerobic conditions, nitrogen (N) is lost as N_2O during the nitrification of ammonium (NH_4^+) and during the denitrification reduction of nitrate (NO_3^-) to nitrogen (N_2). Prevailing anaerobic conditions during the denitrification process leads to the production of N_2O as an intermediate product [15], which contributes to an increase in net GHG emissions as CO_2-eq from rice paddies.

Rice is a staple food for over half of the global population, yet the greenhouse gas intensity (GHGi, expressed as kg CO_2-eq kg^{-1} grain yield) of rice is reported to be the highest among the major crops, because of high CH_4 and N_2O emissions from rice [13,16,17]. To meet the growing demand for food for an ever-increasing global population, the use of N fertilizers in rice cultivation has been projected to rise in the coming years, thus contributing towards further GHG emissions [18,19]. Crop management practices such as efficient water and sustainable nutrient management may reduce the need for additional fertilisers [20]. Water management techniques such as, intermittent drainage [21,22] and alternate wetting and drying [23–26], are all recognized as promising methods for mitigating CH_4 emissions. These practices enhance soil permeability and elevate the soil redox potential, which suppresses methanogenesis and consequently reduces CH_4 emissions [27], although there can be trade-offs with N_2O emissions [28]. use of enhanced efficiency fertilizers (EEFs) can mitigate CH_4 emissions from rice cultivation [11].

Enhanced efficiency fertilizers are made by modifying conventional fertilizers with nitrification and urease inhibitors (UI), which can effectively mitigate GHG emissions, particularly N_2O [11,29]. These inhibitors help slow urea's breakdown into ammonium (NH_4^+) and nitrate (NO_3^-) forms, thereby reducing the potential for N_2O emissions [30]. Chemical nitrification inhibitors (NI), such as dicyandiamide (DCD) 3,4-dimethylpyrazole phosphate (DMPP), and nitrapyrin [2-chloro-6-(trichloromethyl) pyridine] are known to reduce N_2O emissions and improve soil enzymatic and nutrient availability under rice cultivation [31–34]. However, natural NIs (NNI) such as neem (*Azadirachta indica*), karanjin (*Pongamia glabra,* Vent.), mahua (*Madhuca longifolia*, L.), and mint (*Mentha spicata, Mentha arvensis* L.) have been known to inhibit effectively the activities of the nitrifiers [11,35,36]. It has been reported that neem oil coated urea (NCU) and 0.1 % of karanj oil coated urea (KCU) could reduce CO_2-eq emissions by 18.6 % and 21.5 % respectively from rice [11]. Increasing the concentration of NIs contributes to further reduction in GHG emissions from rice [37,38].

Slow-release fertilisers (SRFs) are a type of EEF, which provide a steady supply of nutrients over an extended period. This reduces the need for frequent fertilizer applications [39], minimizes N leaching and subsequent emissions [40] and decreases nutrient losses through leaching and volatilization. In addition, SRFs also support soil health by stimulating the microbial biomass [41,42]. Several well know SRFs such as biochar based SRF and polymer based SRF have shown promising results in reducing GHG emissions, improved nutrient use efficiency (NUE), soil health and rice yield [43–45]. Among these, sulphur coated urea (SCU) has also shown comparable performance across the same metrics. Nutrients from SCU are gradually released via the micropores in the coating material and the fissures that appear after the sulphur coating breaks down [45], thereby improving the morphological and yield characteristics of rice through enhanced N uptake [46,47]. In addition to improving yield and NUE, 5 % coating of bentonite sulphur on prilled urea has demonstrated an improvement in returns and the cost-benefit ratio for aromatic rice in the western IGPs [48]. Selecting appropriate fertilizers and inhibitors is crucial in reducing GHG emissions in paddy fields. Due to rice's unique oxygen limited environment, denitrification of the applied N becomes particularly important, leading to potentially large N_2O emissions from wet paddy rice systems [49].

Soil health, vital for ecosystem recovery, depends on a balanced mix of chemical, physical, and biological components, with enzymes playing a key role [50]. Although soil organic carbon (SOC) is an important indicator of soil quality, it responds very slowly to changes in organic carbon or nitrogenous inputs [51]. Microbial biomass carbon (MBC) and microbial biomass nitrogen (MBN) are critical indicators of soil quality as they represent the active part of soil organic matter. They are involved in nutrient transformations and storage, and their rapid turnover makes them sensitive indicators of changes in soil management, climate, and pollutant toxicity. High levels of MBC and MBN indicate a healthy and active microbial community, essential for nutrient cycling and soil fertility [52]. Dehydrogenase activity (DHA) is a key indicator of biological activity in soils, reflecting the oxidative processes essential for soil health. This enzyme is integral to soil microorganisms' respiration pathways and facilitates the oxidation of organic matter by transferring protons and electrons. Monitoring DHA helps assess the soil's potential to support essential biochemical processes, thereby indicating soil fertility and overall health [50]. Nitrate reductase activity (NRA) catalyzes the conversion of nitrate (NO_3^-) to nitrite (NO_2^-) in the denitrification process. It reflects the efficiency of nitrogen cycling and potential impact of fertilizers and pesticides on soil microbial populations and denitrification potential [53]. On the other hand, urease enzyme converts urea into ammonium (NH_4^+) and plays a crucial role in nitrogen cycling. Its activity reflects the soil's capacity to utilize urea-based fertilizers effectively [54]. These enzymatic processes influence soil nitrogen transformations and microbial respiration, which are key pathways for GHG emissions and nutrient availability [55,56]. Thus, assessing enzyme activity offers valuable insight into soil fertility and environmental performance of fertilization practices.

Since 2015, the Government of India (GoI) has enforced a policy requiring the production and distribution of NCU [57] for agricultural consumption, which has entirely replaced availability and use of PU. However, neem availability may be limited in other countries, while opportunities for further improvement exists, and other alternate natural inhibitors need to be explored. As part of initiatives for promotion of alternate fertilizers, SCU also called as 'Urea Gold' has recently been launched by the GoI to address the issue of S deficiency in the soil. While many studies have measured GHG emissions from rice systems, few have compared multiple enhanced efficiency fertilizers (EEFs) for their combined effects on greenhouse gas intensity (GHGi) and soil health. This study addresses that gap by evaluating the comparative performance of EEFs: neem-coated urea (NCU), karanj-coated urea (KCU), and sulphur-coated urea (SCU) in transplanted rice cultivated in the Indo-Gangetic Plain (IGP) region. Beyond emissions, we assessed their impact on microbial biomass, nutrient availability, and key enzymatic indicators of soil health, including dehydrogenase (DHA), nitrate reductase (NRA), and urease activity. We also projected their mitigation potential at the regional scale, using field-based emission data from two consecutive years to estimate GHGi reductions under rice cultivation in the intensively cultivated rice-wheat cropping system in the IGP. The study offers direct field-based evidence to frame sustainability strategies in rice cultivation under the changing climatic scenario.

2. Materials and methods

2.1. Site description

The field experiment was carried out over the kharif (rainy season) periods of 2019 and 2020, by growing the Pusa 44 rice variety. The experimental farm, situated at the Indian Agricultural Research Institute in New Delhi, India, is positioned in the Indo-Gangetic alluvial tract at 28°40′ N and 77°12′ E, at an elevation of 228 m above mean sea level. The region has a subtropical and semi-arid climate, receiving an annual rainfall of 750 mm, with approximately 80 % occurring between July and September. During the July to October period, mean maximum and minimum temperatures were 35 °C and 18 °C, respectively. The soil at the experimental site had a bulk density of 1.4 g cm^{-3}, electrical conductivity of 0.37 dS m^{-1}, pH (1:2 soil:water) of 8.02, cation exchange capacity of 4.8C mol (p^+) kg^{-1}, total N concentration of 0.30 g kg^{-1}, Olsen P at 0.0078 g kg^{-1}, organic carbon content of 4.2 g kg^{-1} and ammonium acetate extractable K at 0.13 g kg^{-1}. Meteorological parameters including rainfall, temperature and relative humidity were recorded from the nearby meteorological (Fig. S1).

2.2. Experimental design and treatments

The field was ploughed and flooded 2–3 days before transplanting for puddling and levelling. Rice plants (30-day seedlings of cv. Pusa 44) were transplanted at a spacing of 20 × 15 cm in the field plots (6 m × 7 m) and grown in an intermittent flooding regimen. The experiment was laid out in a randomized block design, with five distinct nitrogen treatments, each comprising three replicates. A total of fifteen and sixteen irrigations were applied in 2019 and 2020 respectively, when fine cracks developed on the soil surface. Each irrigation was systematically applied at 5–6 cm of water depth throughout the growth period using the check basin technique. Comprehensive measures were implemented to manage and control weeds, diseases, and pests to ensure the integrity and health of the experimental crop.

The five treatments were (i) control (Con): without fertilizer amendment, (ii) prilled urea (PU), (iii) neem coated urea (NCU) (iv) karanj coated urea (KCU)and (v) sulphur coated urea (SCU). The nitrogen application rate was consistent across different fertilizers, at 120 kg N ha^{-1}. Transplanting was carried out on July 25, 2019 and August 3, 2020 respectively. All the fertilizers were applied through three split doses, where 50 % was applied at 15 days after transplanting (15 DAT), 25 % at maximum tillering (45 DAT) and 25 % at flowering (65 DAT). A basal dose of single super phosphate (26 kg P ha^{-1}), muriate of potash (50 kg K ha^{-1}) and zinc sulfate (10 kg Zn ha^{-1}) was applied to the soil before transplanting.

2.3. Preparation of coated urea fertilizers

Commercially available NCU was used (supplied by Indian Farmers Fertiliser Cooperative (IFFCO)) with 350 ppm of neem oil over prilled urea. For this study, KCU was prepared by coating 100 g PU with 1 % Karanj oil emulsion (oil + hexane), as described by Sahrawat [58]. Similarly, SCU was also prepared specifically for the study, by mixing 950 g of PU with 2 g of gum acacia and 50 g of finely powdered bentonite sulphur (90 % to pass 100 mesh sieve) as described by Shivay et al., [47]. The coated urea thus contained 5 % sulphur by weight.

2.4. Plant sampling and estimation of yield

Plant samples were collected at different crop growth stages, viz. maximum tillering, flowering and at maturity. Yield parameters, such as total biomass and grain yield were recorded after the final harvest from one square meter area of each plot in triplicate. The grains were separated from the straw, dried and weighed. Grain moisture was determined immediately after weighing, and sub-samples were dried in an oven for 48 h at 65 °C. The dried grain and biomass samples were ground and used to estimate the N content using the Kjeldahl method [59]. Total N uptake was calculated from N content in grain and straw. The additional effect of fertilizer application on biomass and yield over the No N plots were reported as apparent recovery (AR) (Eq. (1)) and agronomic efficiency (AE) (Eq. (2)), i.e.,

$$AR(\%) = \frac{N\,uptake\,for\,N\,fertilized\,plot\,(kg\,ha^{-1}) - N\,uptake\,for\,N_0\,plot\,(kg\,ha^{-1})}{Applied\,N\,in\,N\,fertilized\,plot\,(kg\,ha^{-1})} \quad (1)$$

$$AE\,(kg\,grain\,kg^{-1}\,N\,applied\,ha^{-1}) = \frac{Grain\,yield\,in\,N\,fertilized\,plot - Grain\,yield\,in\,N_0\,plot}{Applied\,N\,(kg\,ha^{-1})} \quad (2)$$

where N_0 plot is unfertilized control.

2.5. Collection and analysis of gas samples and fluxes

Greenhouse gas fluxes from the rice field plots were measured using the static chamber method [60]. Three flux chambers were installed in each treatment. Air samples for analysis of CH_4 and N_2O were collected from acrylic chambers of 50 cm × 30 cm x 100 cm (length x width x height), which enclosed the rice canopy and soil surface. The chambers were equipped with a thermometer, a battery-operated fan and a rubber septum to facilitate sampling. An aluminium base frame (channel) of 15 cm height and 5 cm internal diameter was inserted to a depth of 10 cm in soil. Water was filled in the channel to ensure an airtight seal. Gas sampling was conducted weekly between 9 and 11 a.m., using a 50 ml syringe with a three-way stopcock at 0, 30 and 60 min after the closure of the chamber and filled into 10 ml pre-evacuated exetainers. Additionally, air samples were collected on both 0 and 1 days after transplanting (DAT), followed by regular seven-day intervals.

Samples were analyzed for CH_4 and N_2O using a gas chromatograph fitted with a flame ionization detector (GC 8 A, Shimadzu) and electron capture detector (Hewlett Packard 5890 Series II) respectively. Cumulative CH_4 and N_2O emissions were calculated by linear interpolation between two successive measurement intervals on the sampling days. A linear trend of GHG emissions was assumed during the periods when no samples were collected for the calculations [61].

The emissions of CH_4 and N_2O from soil were calculated using Eq. (3):

$$F = \rho \times \left(\frac{V}{A}\right) \times \left(\frac{\Delta c}{\Delta t}\right) \times \left(\frac{273}{T}\right) \quad (3)$$

where F is the CH_4/N_2O flux (mg CH_4 m^{-2} h^{-1}/μg N_2O m^{-2} h^{-1}), ρ is the gas density, V is the volume of chamber (m^3), A is the surface area enclosed by the gas chamber (m^2), Δc/Δt is the rate of increase of CH_4/N_2O gas concentration in the chamber (mg/μg m^{-3} h^{-1}) and T (absolute temperature) was calculated as 273 + mean temperature in °C of the chamber [8].

The total CH_4/N_2O flux was calculated for the entire cultivation period using Eq. 4

$$Total\ gas\ flux = \sum_{i}^{n} (R_i \times D_i) \quad (4)$$

where, R_i is the CH_4/N_2O emission flux (g m^{-2} d^{-1}) on the i^{th} sampling interval, D_i is the number of days in the ith sampling interval, and n is the number of sampling intervals [62].

2.6. Estimation of CO_2-eq and greenhouse gas intensity

The CO_2-eq emission was calculated using global warming potential of 21 and 310 respectively [63] (Eq. (5)) and greenhouse gas intensity (GHGi) was calculated using Eq. (6) [64].

$$CO_2 - eq\ emission\ (kg\ ha^{-1}) = seasonal\ CH_4\ emission\ (kg\ CH_4\ ha^{-1}) \times 21 + seasonal\ N_2O\ emission\ (kg\ N_2O\ ha^{-1}) \times 310 \quad (5)$$

$$GHGi(kg\ CO_2 - eq\ kg^{-1}\ grain\ yield) = \frac{CO_2 - eq\ (kg\ ha^{-1})}{grain\ yield\ (kg\ ha^{-1})} \quad (6)$$

2.7. Soil sampling and analysis

Soil samples were collected in triplicates from different treatment plots (0–15 cm) during different growth stages of the rice. Composite soil samples were prepared by manually removing stones and root fragments. The soil moisture content was determined gravimetrically.

2.8. Nitrate-N and Ammoniacal-N

Soil samples were collected during each gas sampling events for the analysis of nitrate-N and ammoniacal-N. Nitrate-N and Ammoniacal-N were measured by colorimetric methods using N flow auto analyser (Skalar San++). Soil samples were extracted with 2M KCl solution (1:2 soil: extractant) and the extracts were analyzed for ammoniacal-N and nitrate-N using modified Berthelot method [65] and cadmium reduction method (ISO 13395) [66], respectively. The pH of the ponding water was measured on the 3rd and 10th days following each application of split doses of N fertilizer.

2.9. Soil microbial biomass carbon and microbial biomass nitrogen

The soil microbial biomass carbon (MBC) and microbial biomass nitrogen (MBN) were estimated by chloroform fumigation-extraction method (CFE) by Jensen and Sorensen [67] and Brookes et al., [68] respectively. Water saturated soil samples were mixed with two ml methanol free chloroform. The resulting soil suspension was thoroughly mixed and incubated for 24 h in the dark at 25 °C. Subsequently, fumigated and unfumigated samples were extracted using 0.5 M K_2SO_4 solution and filtered. The filtrates were tested for MBC and MBN by titration method. The extraction efficiency coefficient k_{EC} of 0.25 ± 0.05 [69] and 0.45 ± 0.05 [70] were used to measure MBC and MBN respectively.

2.10. Soil enzyme activities

Dehydrogenase activity (DHA) in soils was assessed according to the methodology outlined by Klein et al., [71]. To measure the DHA, 5 g soil sample was mixed with 50 mg $CaCO_3$ and 1 ml of 3 % (w/v) 2,3,5-triphenyl-tetrazolium chloride (TTC) solution and incubated at 37 °C for 24 h. The dehydrogenase enzyme converts TTC to 2,3,5-triphenylformazan (TPF). Acetone extracted TPF was filtered through Whatman No. 42 paper and the optical density was measured at 485 nm with a UV–Visible spectrophotometer. The nitrate reductase activity (NRA) was determined by the method outlined by Keeney and Nelson (1982). To measure the NRA, 5 g of fresh soil was treated with 10 mL of 500 mg NO_3^--N solution and incubated at 28 °C for 24 h. After incubation, 40 mL of 2.5M KCl was added, and the mixture was shaken for 1 h. The filtered extract was then reacted with diazotizing and coupling reagents, and the colour intensity was measured at 540 nm using a spectrophotometer. The NRA was calculated based on the NO_2^--N content determined from a calibration graph. The urease activity were determined by the methods outlined by Bremner and Douglas, [72]. For the measurement, 5 g of fresh soil was treated with 5 mL of urea solution and incubated at 37 °C for 5 h. After incubation, 50 mL of 2M KCl-PMA solution was added and shaken for 1 h. The filtered extract was reacted with diacetylmonoxime and thiosemicarbazide, boiled for 30 min, then cooled. The absorbance was measured at 527 nm using a spectrophotometer, and urease activity was calculated based on urea content from a calibration graph.

Table 1
Total biomass, grain yield and total N as observed under different fertilizer treatment (mean and standard deviation of three replicate plots of each treatment).

Treatment	Total Biomass (kg ha^{-1})			Grain Yield (kg ha^{-1})			Total N uptake (kg ha^{-1})		
	2019	2020	Mean	2019	2020	Mean	2019	2020	Mean
Con	8900 ± 119.8 d	10040 ± 151.0 c	9470 ± 806.1 a	3060 ± 49.1 d	3210 ± 65.6 c	3130 ± 108.3 c	64.0 ± 1.01 e	67.3 ± 0.77 d	65.6 ± 2.34 c
PU	13360 ± 108.1 c	13500 ± 240.0 b	13430 ± 98.9 b	4840 ± 60.0 c	5180 ± 65.1 b	5010 ± 107.3 b	113.4 ± 1.17 d	114.0 ± 0.73 c	113.7 ± 0.43 b
NCU	13720 ± 82.3 bc	14300 ± 151.0 a	14010 ± 410.1 b	5190 ± 39.3 b	5530 ± 125.3 a	5360 ± 140.4 a	118.8 ± 0.78 c	120.1 ± 1.64 b	119.4 ± 1.03 ab
KCU	14590 ± 195.5 a	14500 ± 346.4 a	14545 ± 63.6 b	5470 ± 65.7 a	5650 ± 55.1 a	5560 ± 129.4 a	129.5 ± 2.39 a	129.0 ± 1.61 a	129.2 ± 0.32 a
SCU	13780 ± 207.5 b	14440 ± 302.0 a	14110 ± 466.7 b	5500 ± 25.2 a	5690 ± 72.3 a	5600 ± 134.4 a	123.3 ± 2.11 b	128.7 ± 2.04 a	126.0 ± 1.81 a

Means with at least one letter in common are not statistically significant using Tukey's honest significant difference (HSD). Con = control; PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

2.11. Soil quality index

To better reflect soil health, soil quality index (SQI) was calculated to provide a comprehensive assessment of different treatments. To ensure comparability, all the soil parameters (NRA, urease, DHA, MBC, MBN, NO_3^--N and NH_4^+-N) were first normalized using a min-max approach (Eq. (7)) [73].

$$x' = \frac{x - x_{\min}}{x_{\max} - x_{\min}} \tag{7}$$

where, x′ is the normalized value of the variable, x is the original measured value of the variable, x_{min} and x_{max} are the minimum and maximum observed values of that variable across all treatments respectively.

After normalization of individual indicators, SQI was computed using two common approaches, *i.e.*, equal weight method and PCA-weighted method. However, since the equal weight approach assumes all indicators contribute equally to the index, it ignores the potential inter-variable correlations, thus producing misleading results [74]. Therefore, only PCA-weighted SQI (Eq. (8)) was used for the further calculations and interpretation as it assigns weights based on principal component loadings [75].

$$SQI\text{-}PCA = \sum_{i=1}^{n} x'_i . w_i \tag{8}$$

where, n is the number of variables integrated in index, w_i is the weight (loading) of the *i*th variable.

2.12. Estimated GHG and projected GHGi mitigation potential

The soil used in this study is representative of the typical conditions found in the Indo-Gangetic Plain (IGP), sharing similar physicochemical properties, cropping patterns, and management practices common across the region. Given this representativeness, the results from our experimental plots were used to make broader projections for rice cultivation in the Rice-Wheat Cropping System (RWCS) in the IGP region, which spans approximately 10.5 million hectares [11]. To extrapolate the findings, N_2O and CH_4 emissions, along with GHGi under PU, NCU, KCU and SCU, were calculated using Eqs. (9)–(11). These calculations were based on the emission data collected from the field experiments, providing an estimation of the potential GHG mitigation across the RWCS in the IGP with large-scale adoption of EEFs.

$$N_2O\ Emission\ (Gg) = \frac{Fertilized\ area\ under\ rice\ cultivation\ in\ IGP\ (ha) \times Mean\ N_2O\ (kg\ N_2O\ ha^{-1})}{10^6} \tag{9}$$

$$CH_4\ Emission\ (Gg) = \frac{Fertilized\ area\ under\ rice\ cultivation\ in\ IGP\ (ha) \times Mean\ CH_4\ (kg\ CH_4\ ha^{-1})}{10^6} \tag{10}$$

$$GHGi\ (Gg\ CO_2\ eq.\ kg^{-1}\ grain\ yield) = \frac{Fertilized\ area\ under\ rice\ cultivation\ in\ IGP\ (ha) \times Mean\ GHGi\ (kg\ CO_2\ eq.\ kg^{-1} grain\ yield)}{10^6} \tag{11}$$

For the projections of the effectiveness of EEFs, mean N_2O, CH_4 emissions and GHGi were considered from both the years. Application rate of the EEFs were assumed as 120 N kg ha^{-1}. The reduction percentage was calculated with respect to PU using Eqs. (12)–(14),

$$N_2O\ Reduction\ (\%) = 1 - \frac{N_2O\ emission\ (EEFs)}{N_2O\ emission\ (PU)} \times 100 \tag{12}$$

$$CH_4\ Reduction\ (\%) = 1 - \frac{CH_4\ emission\ (EEFs)}{CH_4\ emission\ (PU)} \times 100 \tag{13}$$

$$GHGi\ Reduction\ (\%) = 1 - \frac{GHGi\ (EEFs)}{GHGi\ (PU)} \times 100 \tag{14}$$

2.13. Benefit cost ratio

The benefit cost ratio of all the treatments was computed to identify the factors responsible for differences in economic benefit associated with each of them. Total grain and straw yield were estimated followed by cost benefit analysis. The current market prices of all the inputs used and hired services during the respective cropping seasons were considered, and these prices were obtained through market research (Tables S2 and S3). Total cost of cultivation, cost of energy, gross income and net return were calculated using Eqs. S1, S2, S3 and S4 respectively. The benefit cost ratio was calculated with gross income and total cost of cultivation using Eq. (15) [76].

$$Benefit\ cost\ ratio\ (B:C) = \frac{Gross\ income\ (Rs)}{Total\ cost\ of\ cultivation\ (Rs)} \tag{15}$$

2.14. Statistical analysis

Statistical analysis of the experimental data was performed using SPSS (26.0, USA). One way ANOVA was carried out to check the statistical significance of the variations between the means. If the ANOVA was found to be statistically significant at 5 % level of significance and the error variances were uniform, Tukey's post hoc test was conducted to identify which treatment means exhibited significant differences. All standard deviations are reported as ± in this study.

3. Results

3.1. Total biomass, grain yield and total N uptake

A significant increase in total plant biomass and grain yield was

Table 2
Apparent recovery (AR) and agronomic efficiency (AE) as measured under different fertilizer treatments (mean of three replicate plots of each treatment).

Treatment	AR (%)			AE (kg grain kg N applied^{-1})		
	2019	2020	Mean	2019	2020	Mean
PU	41.17 ± 0.97 d	38.93 ± 0.61 c	40.05 ± 1.43 c	14.87 ± 0.50 c	16.39 ± 0.54 b	15.63 ± 0.95 b
NCU	45.10 ± 0.67 c	45.92 ± 1.37 b	45.51 ± 1.07 b	18.47 ± 0.50 b	19.19 ± 0.88 a	18.83 ± 0.76 a
KCU	54.57 ± 1.99 a	51.44 ± 1.34 a	53.00 ± 2.29 a	20.11 ± 0.55 a	20.36 ± 0.46 a	20.24 ± 0.47 a
SCU	49.44 ± 1.76 b	51.17 ± 1.87 a	50.31 ± 1.88 ab	20.39 ± 0.21 a	20.69 ± 0.60 a	20.54 ± 0.44 a

Means with at least one letter in common are not statistically significant using Tukey's honest significant difference (HSD). PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

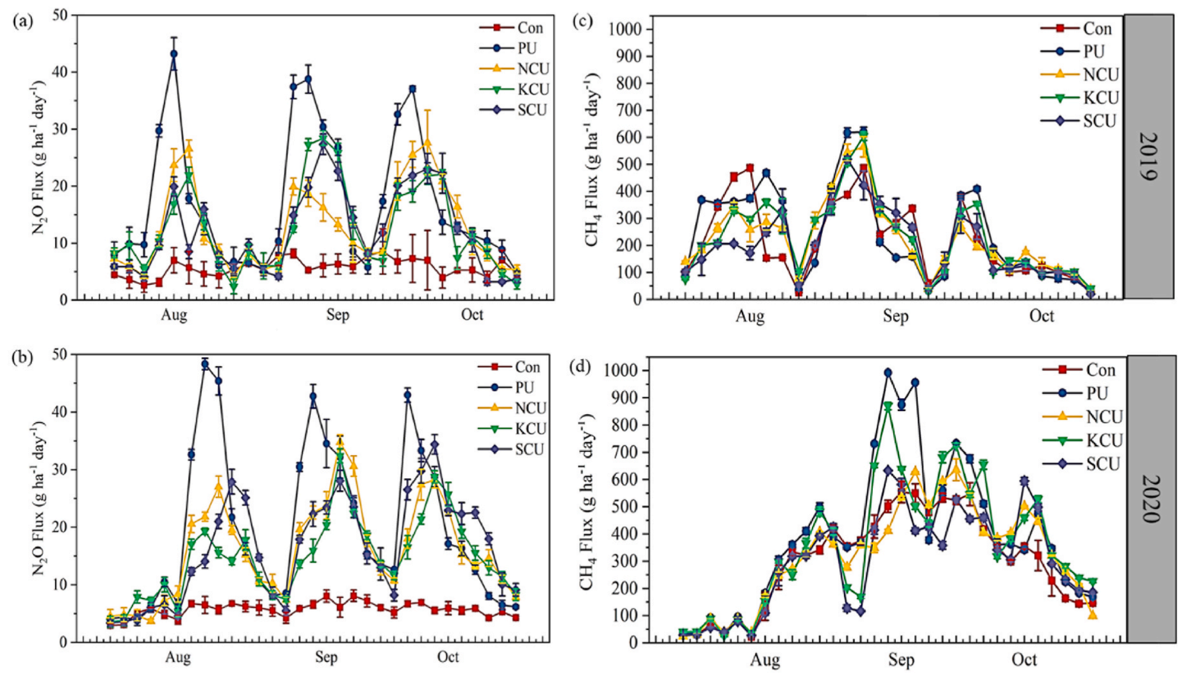

Fig. 1. Mean daily fluxes of (a) N_2O in 2019, (b) N_2O in 2020, (c) CH_4 in 2019 and (d) CH_4 in 2020 are presented in g ha^{-1} day^{-1}. Con = control; PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

observed in plots treated with urea in both years compared to the control treatment (Con). In the 2019 trials, total biomass (grain, straw) in the treated soils ranged from 13360 to 14590 kg ha^{-1}, while in the 2020 trials, the total biomass ranged from 13500 to 14500 kg ha^{-1} (Table 1). The application of coated urea led to a significant increase in total biomass compared to PU. In both the years, KCU had the highest total biomass 14590 kg ha^{-1} and 14500 kg ha^{-1} respectively. The total biomass in NCU and SCU were comparable in both the years. Total biomass under KCU showed no significant difference between 2019 and 2020, whereas both NCU and SCU showed a significant increase in biomass in 2020 compared to 2019. Grain yield also increased following the addition of urea in both years. An increase of 38–44 % was observed in both the years (Table 1). As significant increase in grain yield was following on application of EEFs in both the years in comparison to PU. While no significant differences were observed between the EEFs in 2020, KCU and SCU performed better than NCU in 2019. As shown in Table 1, the application of urea led to a significant increase in total N uptake in both years ($p < 0.05$). The total N uptake in KCU and SCU were comparable in both the years. However, no significant differences in mean total N uptake were observed between the NCU, KCU and SCU.

3.2. Apparent recovery and agronomic efficiency

Agronomic Recovery (AR) with the NCU treatment were comparable in both the years. KCU showed the highest AR in 2019 as compared to other treatments ($p < 0.05$) but was not significantly different than SCU in 2020. Overall, KCU recorded ~32 % higher mean AR when compared with PU.

Agronomic Efficiency (AE) in plots with different types of coated urea was significantly higher than PU in both the years (Table 2). AE in KCU and SCU were comparable in both the years and were significantly higher in 2019 as compared with PU and NCU. However, mean AE among the coated urea types showed no significant difference but was significantly higher than PU. No significant differences were observed between the treatment means as seen in Table 2.

3.3. Greenhouse gas flux

The N_2O flux in the control treatment (Con) varied from 2.6 to 8.6 g ha^{-1} day^{-1} in 2019 (Fig. 1a) and 3.0–8.1 g ha^{-1} day^{-1} in 2020 (Fig. 1b). Following urea application in the different treatment plots, the N_2O flux varied from 2.4 to 43.22 g ha^{-1} day^{-1} in 2019 and 3.4–48.34 g ha^{-1} day^{-1} in 2020. The flux increased for a few days immediately following fertilizer application and then decreased gradually. The temporal variation in N_2O flux indicated distinct peaks for PU, while treatments with coated urea exhibited more gradual peaks over time. The cumulative N_2O-N emissions from all coated urea treatments did not vary significantly in both years. The emissions from PU were significantly higher in both 2019 (0.87 kg N_2O-N ha^{-1}) and 2020 (0.72 kg N_2O-N ha^{-1}) than those from the treatments with coated urea (Table 3). The emissions from all the coated urea fertilizers were lower by ~14 % than PU in both the years. No significant differences in emissions were observed between NCU, KCU and SCU in both the years. An enhanced coating of karanj oil (1 %) in KCU compared with 0.1 % KCU did not lead to any significant change in the N_2O-N emissions in both years (Table S1).

Throughout the growing season, CH_4 emissions increased to steadily at key crop growth stages in all treatments. The CH_4 flux in Con varied from 25.1 to 487 g ha^{-1} day^{-1} in 2019 (Figs. 1c) and 22–554 g ha^{-1} day^{-1} in 2020 (Fig. 1d). The CH_4 flux measured from plots receiving urea varied from 16 to 620 (g m^{-2} day^{-1}) in 2019 and 24 to 992 (g m^{-2} day^{-1}) in 2020. The emissions peaked during the period between late August and early September before returning to pre-transplanting levels. The peak emissions were observed at the heading and flowering phases to the beginning of the grain-filling phase. The PU plots had the highest flux peaks in both years, resulting in significantly ($p < 0.05$) greater cumulative CH_4 emissions than all other treatments. The cumulative CH_4 emissions from coated urea treatments varied from 13.6 to 14.9 kg CH_4- C ha^{-1} in 2019 and 14–15.4 kg CH_4- C ha^{-1} in 2020 (Table 3). The application of coated urea treatments significantly reduced CH_4 emissions compared to PU. NCU and KCU significantly reduced CH_4 emissions by 10.7 % and 8.8 % in 2019 and by 11.1 % and 13.5 % in 2020 respectively in comparison to PU. However, the emissions from SCU were 19 % and 21 % lower than PU in 2019 and 2020 respectively.

3.4. CO_2-eq emission and greenhouse gas intensity

It is evident that the contribution from CH_4 dominated the CO_2-eq emissions (Table 3). Even the plots without fertilizer (Con) application contributed substantially to these emissions. However, the magnitude was low when compared to treated plots. The PU treatment had significantly higher emissions, i.e., 891.6 kg CO_2-eq. ha^{-1} in 2019 and 833.4 kg CO_2-eq. ha^{-1} in 2020. The application of coated urea reduced emissions by 12–20 % in 2019 and 8–15 % in 2020. Although all the coated urea treatments showed significant reduction in emissions as compared to PU, SCU had significantly lower emissions in comparison to all other coated urea treatments in 2020.

The GHGi represents the ratio of CO_2-eq per unit of yield. No significant difference in yield-scaled GWP were observed between the Con and PU treatment (Table 3), indicating no effect of urea application. However, the application of coated urea significantly reduced GHGi compared to the Con and PU treatment. SCU demonstrated the lowest mean GHGi in both years, resulting in a mean reduction of 26.0 % compared with PU, which was a significantly larger reduction than achieved by NCU or KCU. Although NCU and KCU also demonstrated significant reduction in GHGi compared with PU, but they were similar to each other in both the years (mean reduction of 19 %).

3.5. Nitrate-N and Ammoniacal-N

It is evident that urea application significantly increased nitrate N in the soil. The application of coated urea slightly decreased nitrate N in soil compared with PU in 2019, However, this reduction was not seen for 2020 which is consistent with their action as nitrification inhibitors (NCU, KCU) or slow-release fertilisers (SCU). However, this reduction was not seen for 2020. The values were higher in 2019 than in 2020 (Fig. 2). Additionally, the peaks of each treatment were delayed in 2020 compared with 2019 (not shown). All the treatments exhibited different trends in amounts of nitrate N in both years. PU (2.8–7.2 kg N/ha) exhibited the most stable trend with minimal variations in 2020. However, in 2019, PU had exhibited significantly higher values and greater fluctuations (as seen by the ranges shown in Fig. 2). SCU (4–9.6 kg N/ha) and NCU (3.2–9.2 kg N/ha) exhibited the most stable trend with minimal variations in 2019. NCU and KCU followed a similar pattern to PU only in 2019. Overall, the trend indicated that PU exhibited slightly

Table 3
Cumulative N_2O and CH_4 emissions, CO_2-eq and GHGi under different treatments (mean of three replicate plots of each treatment±standard deviation).

Treatment	N_2O Emission (kg N_2O-N ha^{-1})			CH_4 Emission (kg CH_4-C ha^{-1})			CO_2-eq (kg ha^{-1})			GHGi (kg CO_2-eq kg^{-1} grain yield)		
	2019	2020	Mean	2019	2020	Mean	2019	2020	Mean	2019	2020	Mean
Con	0.32 ± 0.04 c	0.27 ± 0.01 c	0.30 ± 0.03 c	13.91 ± 0.31 cd	14.15 ± 0.20 d	14.03 ± 0.17 bc	545.2 ± 27.1 c	528.6 ± 4.1 d	536.9 ± 11.7 c	0.178 ± 0.012 a	0.165 ± 0.003 a	0.172 ± 0.010 a
PU	0.87 ± 0.03 a	0.72 ± 0.01 a	0.79 ± 0.11 a	16.73 ± 0.37 a	17.32 ± 0.33 a	17.02 ± 0.42 a	891.6 ± 21.0 a	833.4 ± 16.4 a	862.5 ± 41.1 a	0.184 ± 0.002 a	0.161 ± 0.004 a	0.173 ± 0.016 a
NCU	0.69 ± 0.04 b	0.62 ± 0.02 b	0.66 ± 0.04 b	14.93 ± 0.50 bc	15.39 ± 0.12 c	15.16 ± 0.48 abc	758.7 ± 34.2 b	745.1 ± 14.5 b	751.9 ± 9.6 b	0.146 ± 0.008 b	0.135 ± 0.005 b	0.140 ± 0.008 b
KCU	0.73 ± 0.04 b	0.64 ± 0.02 b	0.68 ± 0.07 b	15.25 ± 0.42 b	16.35 ± 0.34 b	15.80 ± 0.77 ab	783.1 ± 27.2 b	768.5 ± 17.2 b	775.8 ± 10.3 b	0.143 ± 0.004 b	0.136 ± 0.003 b	0.140 ± 0.005 b
SCU	0.70 ± 0.05 b	0.66 ± 0.02 b	0.68 ± 0.03 b	13.56 ± 0.29 d	13.97 ± 0.28 d	13.77 ± 0.29 c	720.5 ± 33.0 b	712.0 ± 19.0c	716.3 ± 6.0 b	0.131 ± 0.006 b	0.125 ± 0.004 c	0.128 ± 0.004 c

Means with at least one letter in common are not statistically significant using Tukey's honest significant difference (HSD). Con = control; PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

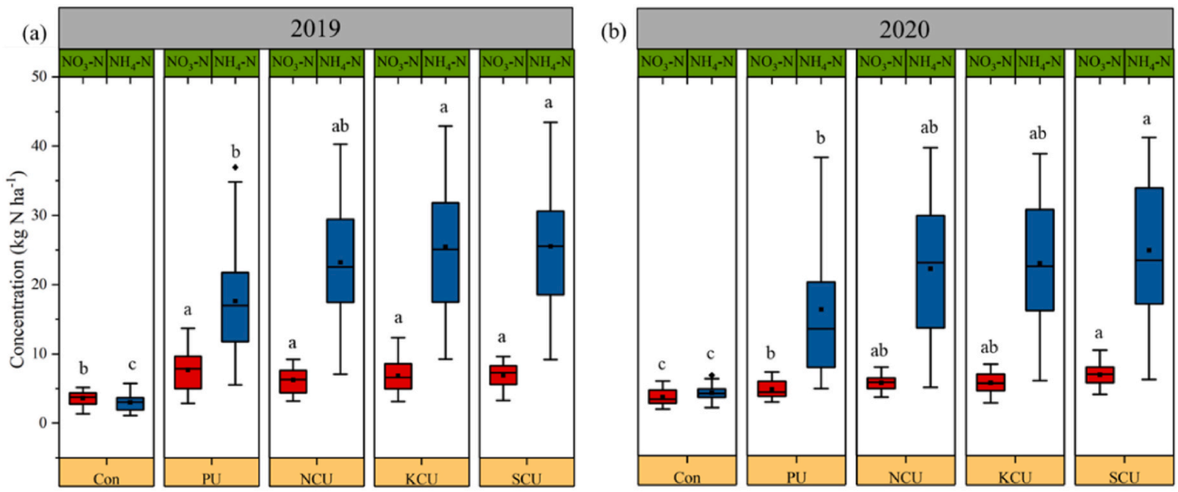

Fig. 2. Soil NO_3^--N (red bars) and NH_4^+-N (blue bars) as measured under different treatments in (a) 2019 and (b) 2020. Con = control; PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

higher nitrate N values than NCU and KCU in 2019 but were slightly lower to them in 2020. SCU exhibited a slightly different variational trend, and lower values compared to PU.

The treated plots demonstrated several times higher ammoniacal-N compared with Con in both the years. Similar to the nitrate N, ammoniacal-N values were higher in 2019 than in 2020 (Fig. 2). The pH of ponding water for PU treated plots ranged from 6.9 to 8, while in NIs it ranged from 6.9 to 8.2, whereas in the SCU treatment it ranged from 6.8 to 7.9 (Fig. S2) up to ten days after each split of N fertilizer application. The application of coated urea enhanced ammoniacal-N in soil compared to PU, consistent with NCU and KCU acting as nitrification inhibitors. The higher value of soil ammonium for SCU compared with PU is surprising given that this is assumed to act as a slow-release fertilizer and suggests that SCU also has activity as a nitrification inhibitor. Additionally, the coated urea showed higher concentration of ammoniacal-N than PU in both the years (Fig. S3). NCU and KCU had slightly higher concentration compared with PU in both years. NCU showed higher concentration compared to KCU in 2020. However, KCU and SCU exhibited a comparable variational trend in both years (Fig. S3).

3.6. Soil microbial biomass carbon and microbial biomass nitrogen

The flowering phase had the highest microbial biomass carbon (MBC) followed by the physiological maturity phase in both years. Although MBC for PU was higher than the control treatment (Con), the difference was not significant in both the years (Fig. 3a). In both the years, a general trend was observed that the application of coated urea significantly increased MBC compared with PU during maximum tillering and flowering phases. Although the application of coated urea also increased MBC compared with PU during the physiological maturity phase, the difference among the coated urea fertilizers were not significant. The application of KCU led to a significantly higher MBC compared to PU at all three stages in both years, it was not significantly different than other treatments. Also, no significant differences were observed between the other coated treatments during all three phases in both years.

The flowering phase showed the highest microbial biomass nitrogen (MBN) followed by the physiological maturity phase in both years, similar to MBC. Contrary to MBC however, MBN in PU-treated plots were significantly higher compared with controls in both years (Fig. 3b). In both years, NCU-treated plots had significantly lower MBN compared with PU at the three growth stages. Contrary to MBC, except for KCU, urea coating reduced MBN in both years at all three stages. The highest MBN was observed for KCU and was significantly higher than compared with other treatments in both years, except PU in the maximum tillering stage. Similar to NCU, SCU also showed significantly lower MBN in comparison to KCU and PU.

3.7. Soil enzyme activities

It is evident that the flowering phase had the highest Dehydrogenase Activity (DHA) followed by the physiological maturity phase in both the years. In both the years, the DHA in PU treatment was significantly higher than Con (Fig. 3c). The application of NCU and KCU had significantly higher activity compared to PU in both the seasons. Application of KCU lead to a 40–50 % increase in DHA as compared to PU during all the three stages in both years. SCU had exhibited significantly higher DHA compared to PU in the years. The treated plots had significantly higher NRA compared to the Con during all three phases in both years. The variation trend was similar to that of DHA, i.e., highest activity in flowering phase followed by the physiological maturity phase in both years (Fig. 3d). The highest activity was observed in plots with uncoated urea (PU). The application of coated urea led to a significant decrease in the NRA compared to PU. This reduction was 23 %–57 % during the maximum tillering stage, 20 %–52 % during the flowering stage and 22 %–54 % during the physiological maturity stage. No significant differences were observed between NCU and KCU in both the years. NRA in SCU were significantly higher than other treatments, but the differences were significant only in 2019 during all the three phases. Consistent with the variation in DHA and NRA, urease activity was highest during the flowering stage, followed by the physiological maturity stage. The results indicated that the urea treatment generally led to significant urease activity during all three phases in both years (Fig. 3e). The highest urease activity was observed in PU, which was significantly higher than the Con and other treatments. The application of coated urea reduced urease activity compared to PU. Significant variations in activity levels were observed across treatments only during specific stages rather than consistently throughout. Among coated urea treatments, SCU exhibited highest urease activity in 2019, while KCU and SCU exhibited highest urease activity in 2020.

3.8. Correlation of greenhouse gases with enzyme activity

The correlation analysis presented as a heatmap in Fig. 4 provides significant insights into the interrelationships between various soil physicochemical parameters, enzymes activities and GHG from the paddy field treated with different coated urea fertilizers. The N_2O flux

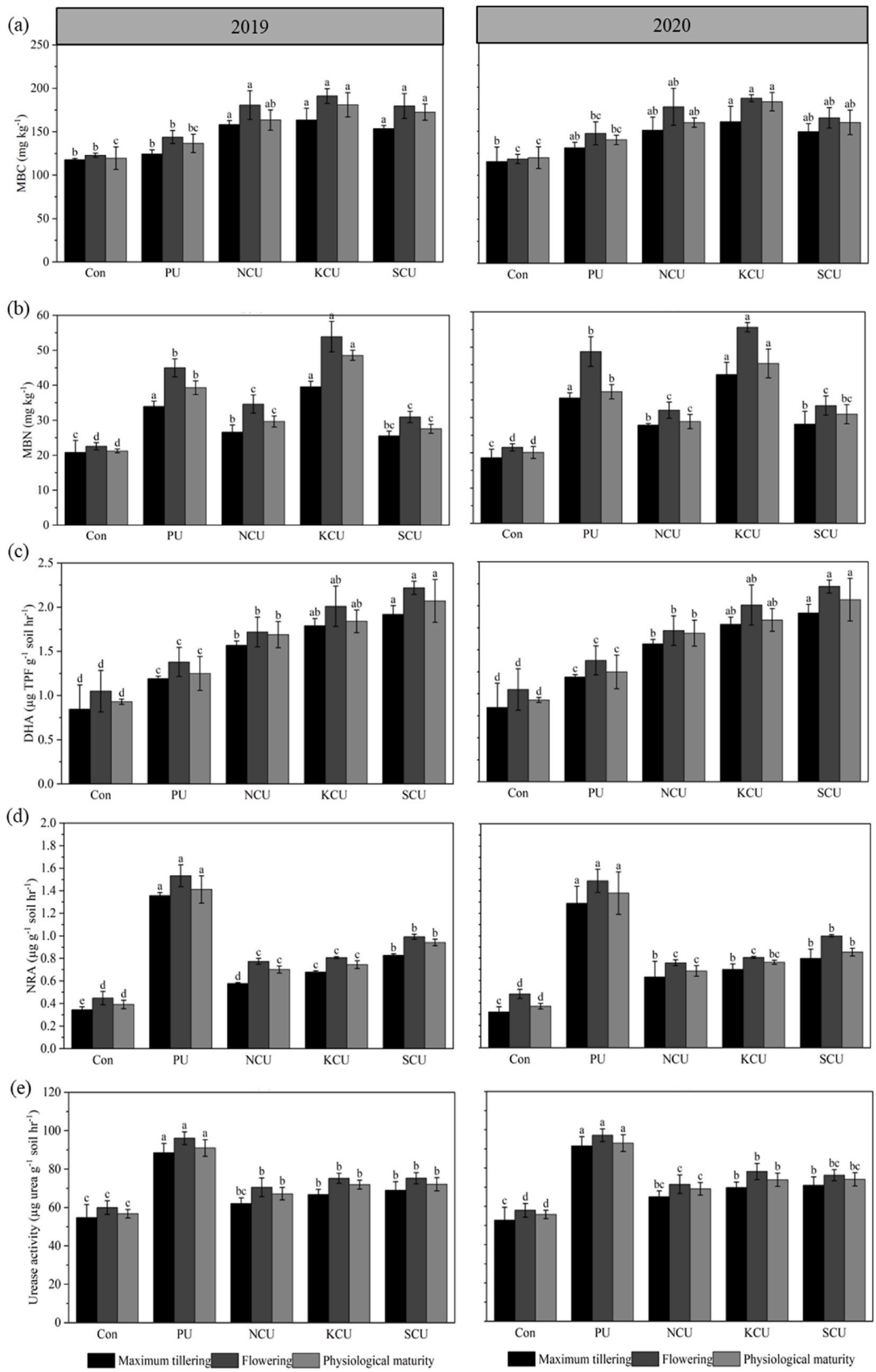

Fig. 3. Soil health indicators: (a) Microbial biomass carbon (MBC), (b) Microbial biomass nitrogen (MBN), (c) Dehydrogenase Activity (DHA), (d) Nitrate Reductase Activity (NRA) and (e) urease activity, as measured under different treatments in 2019 and 2020. Con = control; PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

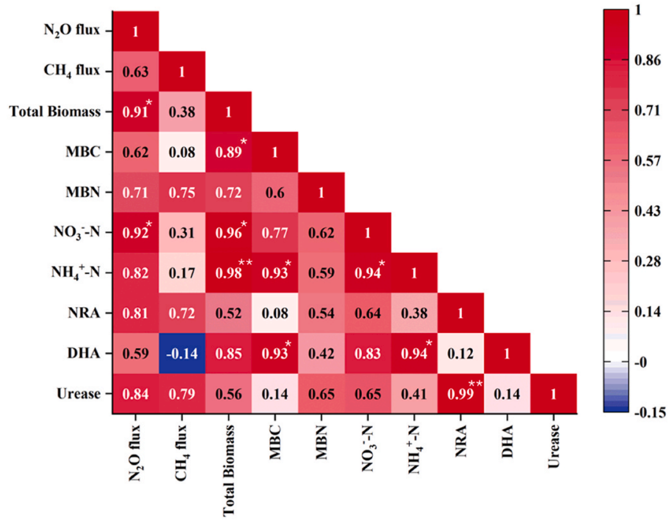

Fig. 4. Heatmap of the relationship between N_2O and CH_4 flux, total biomass and soil microbial, nutrient and enzymatic activities respectively. The color scale on the right ranges from −0.15 to 1, where dark red represents a strong positive correlation (close to 1), white represents no correlation (0), and blue indicates a negative correlation (close to −0.15). * Significant at the 0.05 level, ** significant at 0.01 level.

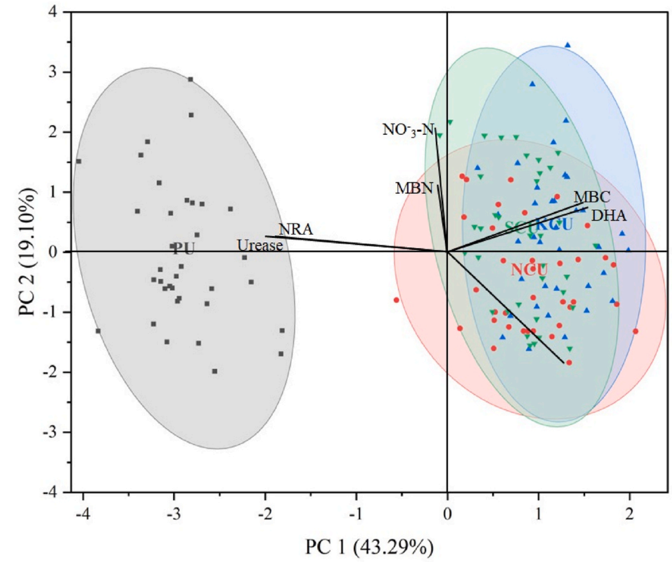

Fig. 5. Principal component analysis (PCA) biplot (PC1: 43.29 %, PC2: 19.10 %) illustrating the separation of treatments based on normalized soil quality parameters and their associations with soil indicators. Symbols represent individual replicates: ■ = prilled urea (PU), ● = neem coated urea (NCU), ▼ = sulphur coated urea (SCU), ▲ = karanj coated urea (KCU); ellipses denote 95 % confidence intervals for each treatment group.

Table 4
Principal Component Analysis-based Soil Quality Index (SQI-PCA) values (mean of replicate plots of each treatment±standard deviation).

Treatment	SQI-PCA		
	2019	2020	Mean
PU	−0.47 ± 0.11 c	−0.44 ± 0.12 c	−0.46 ± 0.02 c
NCU	0.49 ± 0.15 ab	0.42 ± 0.09 ab	0.45 ± 0.05 ab
KCU	0.52 ± 0.08 a	0.51 ± 0.09 a	0.51 ± 0.01 a
SCU	0.41 ± 0.08 b	0.39 ± 0.11 b	0.40 ± 0.01 b

Means with at least one letter in common are not statistically significant using Tukey's honest significant difference (HSD). PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

exhibited strong positive correlations with total plant biomass (r = 0.91), soil NO_3^--N (r = 0.92), soil NH_4^+-N (r = 0.82), and NRA (r = 0.81), indicating that higher N_2O emissions are associated with increased plant or microbial biomass, N content of leaves, and NRA. Conversely, the N_2O flux was negatively correlated with DHA (r = −0.59), suggesting that higher microbial activity may reduce N_2O emissions.

The CH_4 flux showed positive correlations with N_2O flux (r = 0.63), MBN (r = 0.75), NRA (r = 0.72), and urease activity (r = 0.79), and a negative correlation with DHA (r = −0.14), indicating complex interactions between CH_4 emissions and soil microbial processes. Total biomass was strongly positively correlated with soil NO_3^--N (r = 0.96), soil NH_4^+-N (r = 0.98), and DHA (r = 0.87), highlighting the interconnectedness of biomass production and N cycling. Positive correlation was observed between MBC and DHA (r = 0.87) but showed weak correlation with CH_4 flux (r = 0.08). On the other hand, MBN displayed positive correlations with CH_4 flux (r = 0.75) and soil NO_3^--N (r = 0.77), while showing weak correlation with NRA (r = 0.08). Soil NO_3^--N were strongly positively correlated with soil NH_4^+-N (r = 0.94), and N_2O flux (r = 0.92), and positively correlated with DHA (r = 0.83) and urease activity (r = 0.65). Soil NH_4^+-N exhibited strong positive correlations with soil NO_3^--N (r = 0.94), and DHA (r = 0.94), while showing a weak correlation with NRA (r = 0.08). Soil NRA was positively correlated with N_2O flux (r = 0.81) and CH_4 flux (r = 0.72), with weak positive correlations with MBN (r = 0.08) and NH_4^+-N (r = 0.08). Soil DHA showed strong positive correlations with total biomass (r = 0.87) and NH_4^+-N (r = 0.94), while negatively correlating with N_2O flux (r = −0.59). Finally, urease activity demonstrated strong positive correlations with CH_4 flux (r = 0.79) and NRA (r = 0.99), and positive correlations with N_2O flux (r = 0.84) and NO_3^--N (r = 0.65), with weak correlations with MBN (r = 0.41). These findings underscore the intricate and multifaceted relationships between GHG fluxes, soil microbial activities, and nutrient dynamics, which are crucial for developing effective strategies for sustainable soil management and mitigation of GHG emissions.

3.9. Soil quality index and PCA biplot analysis

The PCA weighted soil quality index (SQI-PCA) ranged from −0.47 to 0.52, reflecting both positive and negative contributions of soil parameters to PC1 (Table 4). Higher SQI values were observed in 2019 in comparison to 2020. Among treatments, KCU had the highest SQI values followed by NCU and SCU, whereas PU showed negative scores in both the years. The PCA biplot (PC1 = 43.29 %, PC2 = 19.10 %) revealed that MBC and DHA had the strongest positive loadings on PC1 (Fig. 5). In contrast, urease and nitrate reductase (NRA) had negative loadings. The PCA grouping also revealed a distinct separation of treatments with PU positioned on the negative side of PC1 whereas, SCU, KCU, and NCU treatments were grouped on the positive side.

3.10. GHG emission and GHGi mitigation potential under rice cultivation in rice-wheat cropping system in the IGP

Considering the findings of our experiment which is representative of rice cultivation in the rice-wheat system occupying 10.5 Mha area in the IGP region, CH_4 and N_2O emission, CO_2-eq. and GHGi mitigation potential were calculated under two scenarios of application of EEFs, viz., NI and SRF in rice cultivation under the rice-wheat cropping system in the IGP of India. The estimated GHG emissions and GHGi under PU, NI and SRF are presented in Fig. 6. The order of emissions observed under different treatment is PU > NCU > KCU > SCU. It has been estimated that PU contributes 0.79 kg N_2O-N ha^{-1}, 17.02 CH_4-C kg ha^{-1} and 0.173 GHGi (kg CO_2-eq kg^{-1} grain yield). The results demonstrate that NCU can reduce N_2O emissions by 7.5 % and CH_4 emissions by 5.1 %, while KCU can achieve reductions of 13.8 % and 7.2 % in N_2O and CH_4

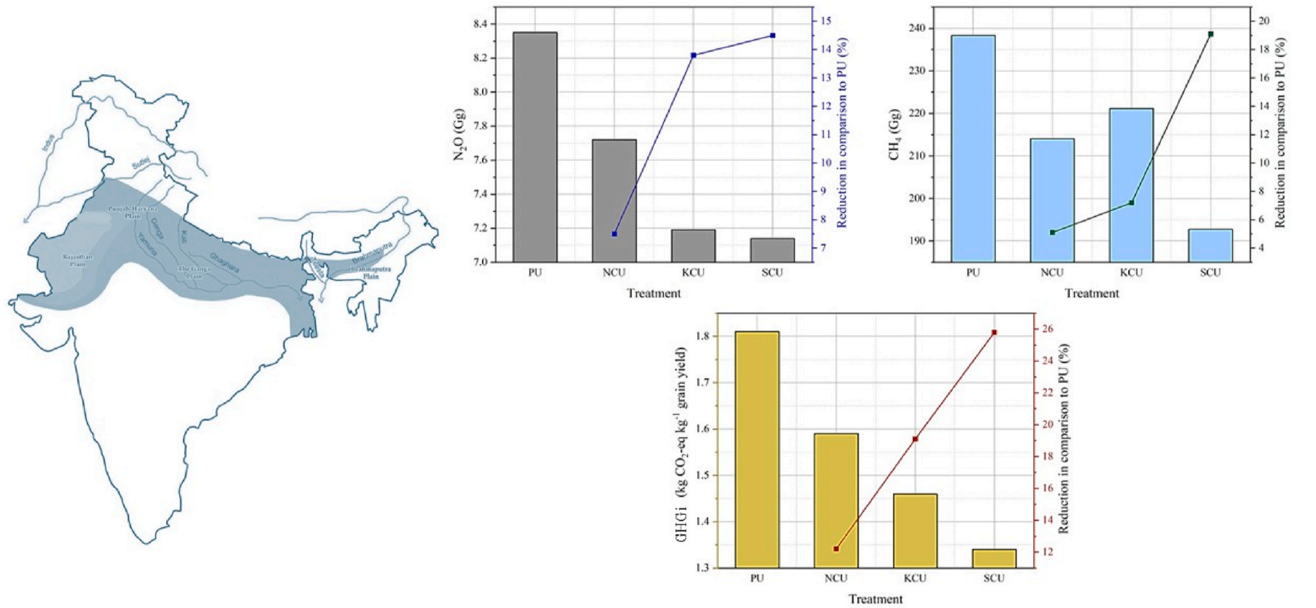

Fig. 6. Predicted greenhouse gas (GHG) emissions (CH_4 and N_2O), greenhouse gas intensity (GHGi), and percentage reduction potential under neem coated urea (NCU), karanj coated urea (KCU) and sulphur coated urea (SCU) in comparison to prilled urea (PU) under rice cultivation in the Indo-Gangetic plains of India.

Table 5
Economic comparison under different treatments.

	Total cost of cultivation (Rs ha^{-1})			Net return (Rs ha^{-1})			Benefit: Cost		
Treatment	2019	2020	Mean	2019	2020	Mean	2019	2020	Mean
Con	57069	59846	58458 ± 1964	16830	20340	18585 ± 2482	1.29	1.34	1.32 ± 0.04
PU	64729	66903	65816 ± 1537	52157	62493	57325 ± 7309	1.81	1.93	1.87 ± 0.08
NCU	65751	67926	66839 ± 1538	61519	69714	65617 ± 5795	1.94	2.03	1.99 ± 0.06
KCU	66169	68344	67257 ± 1538	65931	72793	69362 ± 4852	2.00	2.07	2.04 ± 0.05
SCU	65926	68101	67014 ± 1538	66447	73584	70016 ± 5047	2.00	2.07	2.04 ± 0.05

Con = control; PU = prilled urea; NCU = neem coated urea; KCU = karanj coated urea; SCU = sulphur coated urea.

emissions respectively. On the other hand, application of SCU can reduce N_2O and CH_4 by 14.5 % and 19.1 % respectively. Furthermore, the GHGi can be reduced by 12.2 %, 19.1 % and 25.8 % by NCU, KCU and SCU respectively when compared with PU.

3.11. Economic performance

The cost of cultivation, net return, and benefit: cost (B:C) ratio varied among treatments across both years (Table 5). In 2019, the highest net return (Rs 66,447 ha^{-1}) and B:C ratio (2.00) were recorded in the SCU treatment, closely followed by KCU (Rs 65,931 ha^{-1}; B:C 2.00). Similar trends were observed in 2020, where SCU again achieved the highest net return (Rs 73,584 ha^{-1}) and B:C ratio (2.07), matching that of KCU (Rs 72,793 ha^{-1}; B:C 2.07). The Con treatment consistently showed the lowest net return and B:C ratio in both years (Rs 16,830 and 1.29 in 2019; Rs 20,340 and 1.34 in 2020). Both the NIs (NCU and KCU) outperformed PU and the control, reflecting improved profitability.

4. Discussion

4.1. Crop yield and nutrient efficiency

Total biomass (grain + straw yield) and grain yield were slightly higher in 2020 than 2019, yet both years fell within the anticipated range for the field location [11,77]. The importance of N for crop growth and biomass yield is highlighted by the lower yields observed in the control (Con) plots, emphasizing the fact that supplying additional N is crucial for enhancing rice yields due to the soil's insufficient N supply [78]. Application of neem coated urea (NCU) saw an increase in total biomass and grain yield in comparison with prilled urea (PU) in both years at the same N level. Additionally, total N content, apparent N recovery (AR) and agronomic efficiency (AE) were also significantly higher for NCU than for PU. The improved performance of NCU compared with PU is attributed to its effect as a nitrification inhibitor, resulting in increased soil NH_4^+-N concentrations, which are less liable to be lost than soil NO_3^--N, thereby allowing increased nitrogen recovery. The results obtained on application of NCU support the findings of previous studies [11,79,80]. Efficient N application and its strategic distribution throughout crop growth period are crucial for optimal N utilization and increased yield. Applying NCU can significantly improve dry matter accumulation and yield components, as reflected in enhanced N uptake, use efficiency and recovery [80,81].

In our previous study [11], a 0.1 % concentration of karanj coated urea (KCU) resulted in a 5.6 % increase in grain yield compared to PU. Building on these findings, we increased the karanj oil concentration in the present study to 1 % and observed further improvements. The application of 1 % KCU significantly increased total biomass and grain yield compared to both PU and NCU over two years. Additionally, a significant rise in total N uptake was observed compared to PU, which aligned with our previous results [11]. The higher concentration of KCU (1 %) used in the present study also led to significant improvements AE and AR, and total biomass. Similar results were observed by Kumar et al. [82], whereby increasing the concentration of neem oil significantly enhanced the growth, yield parameters, grain yield, nitrogen uptake and efficiency of aromatic rice. The performance indicators pertaining to the use of KCU 0.1 % and KCU 1 % were compared (Table S1).

As with NCU, the observed increase in crop yield with KCU can be attributed its role as a nitrification inhibitor, which conserves soil NH_4^+-N concentrations. This results in higher availability of N to crop plants, leading to improved crop quality and yield [83]. Additionally, the use of nitrification inhibitors (NIs) typically decreases NO_3^--N content in the soil, enhancing aboveground biomass, N uptake, and nutrient efficiency [43]. In the present study, Fig. 2 shows how NCU and KCU increased NH_4^+-N substantially compared with PU in both years, but NO_3^--N only decreased in 2019, but not 2020. The unexpected results for NO_3^--N in 2020 suggest that other interactions are also important in controlling soil NO_3^--N concentrations (e.g. changed partitioning of root uptake of NH_4^+ versus NO_3^--N) [84].

For many indicators, sulphur coated urea (SCU) performed similarly to NCU or KCU (e.g. indicators where they are not significantly different). Nevertheless, for total biomass, grain yield and total N uptake (See Table 1), SCU was significantly higher than for NCU and KCU. The better performance in terms of yield and efficiency of SCU may be attributed to the presence of sulphur ions which are readily provided to the plants by these fertilizers. A suitable binder such as bentonite in SCU can further slowdown urea release, so that SCU has the benefit of a slow release fertilizer, enabling lower losses and higher AR and AE [85]. It has also been suggested that gypsum-sulphur-based material are considered for reducing the dissolution rate of urea [86], thereby resulting in better availability of N to plants leading to an increase in crop yield and quality [87]. Fig. 1 shows evidence that SCU is indeed acting as a slow-release fertilizer, as illustrated by the delayed N_2O emissions compared to PU. Fig. 2 shows that SCU also increased soil NH_4^+-N concentrations compared with PU, which is unexpected if SCU is only acting as a slow-release fertilizer and by supplying sulphur as a nutrient. The increase in soil NH_4^+-N concentrations compared with PU suggests that SCU also has an effect in inhibiting nitrification. Sulphur in SCU might contribute to nitrification inhibition as volatile sulphur compounds, such as carbon disulfide and hydrogen sulfide, are known to retard nitrification by being considerably effective in closed systems. This suggests that decomposition of sulphur compounds in SCU by soil microorganisms may have formed volatile sulphur compounds that might have inhibited nitrification [88]. Further studies considering the possible effects of SCU on the pH of soil microsites adjacent to dissolving fertilizer granules are needed.

Application of PU often fails to meet the N requirements of rice crops at various growth and maturation stages because of its rapid hydrolysis, nitrification and loss, making it hard to synchronise with crop needs [89]. By contrast NCU, KCU and SCU all provided a more controlled release of N than PU in the present study, as indicated by the temporal distribution of N_2O emissions (Fig. 1). Consequently, the AE in PU treatments is significantly lower than in NCU, KCU and SCU (Table 2). Additionally, the rapid dissolution and leaching of conventional urea can lead to N losses through volatilization and runoff, further diminishing its effectiveness and environmental sustainability.

4.2. Environmental impact

Environmental impacts in this study were assessed by quantifying emissions of N_2O and CH_4, by considering the combined CO_2-eq of these as greenhouse gas (GHG) emissions, and by considering the GHG intensity, expressed as CO_2-eq emission per unit crop yield.

The cumulative N_2O-N emissions from the 2019 samples were significantly higher than those from the 2020 samples ($p < 0.05$). Despite this, the recorded emissions for both years were well below the IPCC Tier 1 default factor of 1 % of applied nitrogen being released as N_2O from mineral N fertilizer applications (Table 3) [28,90]. Although no significant differences were observed in the net N_2O flux between the NCU, KCU and SCU, yet they led to a significant reduction in N_2O emissions compared with PU. A ~14 % decrease in N_2O emission was observed on the application of NCU, KCU and SCU over PU. Nitrification inhibitors are known to significantly reduce N_2O emissions by reducing nitrification by slowing the oxidation rate of NH_4^+ and indirectly reducing NO_3^--N availability for denitrification [91].

It has been reported that lipid associates present in the neem seeds, upon their alcohol extraction, slow the conversion of NH_4^+ to NO_3^- via NO_2^-, thus reducing nitrification in soil [92]. More specifically, the meliacin content in neem oil has been found to directly enhance its nitrification inhibition properties, effectively reducing the rate at which NH_4^+ is converted to NO_3^- [93]. This reduced nitrification rate not only diminishes N_2O emissions (Fig. 1) but also can be linked to a decreased population of NO_2^- oxidizers in the soil. A lower population of these microorganisms results in reduced NO_2^- oxidation, thereby decreasing the availability of NO_3^- for denitrification [11,94]. Consequently, this dual mechanism-direct inhibition by meliacin and reduced nitrite oxidizer activity—contributes to the overall lower N_2O emissions observed with NCU application.

The highest N_2O emissions observed with PU application are primarily attributed to greater nitrogen losses and enhanced nitrification-denitrification activity due to poor N recovery [95–97]. There were no significant differences in N_2O emissions between KCU, SCU and NCU; however, the emissions from these treatments were significantly lower than those from PU. This reduction in N_2O emissions from KCU can be attributed to the furan ring in karanjin, which has been identified as responsible for inhibiting nitrification [87], thereby reducing N_2O emissions [83]. The SCU demonstrated a reduction in N_2O emissions similar to NCU and KCU. Part of this effect can be explained by the slow release of urea in SCU, which reduced the N_2O emissions by reducing the substrate for microbial nitrification and denitrification [98], also illustrated by the longer temporal profile of N_2O emissions with SCU than with the other treatments (Fig. 1). As noted, however, the fact that SCU increased soil NH_4^+N concentration, suggests that it may also have been acting as a nitrification inhibitor. Thus, it is not easy to distinguish how much of the improved efficacy with SCU was due to a) its slow-release profile, b) the additional supply of sulphur as a nutrient and c) its possible action also as a nitrification inhibitor. The present results of increased NH_4^+N with SCU are surprising as elsewhere it has been reported that SCUs improves the efficiency of denitrification, by reducing the overall N_2O:N_2 emission ratio [99]. Therefore, further studies are required to elucidate the relative importance of the mechanisms of action of SCU.

The cumulative CH_4 emissions across all the enhanced efficiency fertilizer (EEF) treatments were significantly smaller than for PU. Although the highest emission was observed in PU (17 kg ha^{-1}), NCU and KCU performed similarly. The reduced CH_4 emissions with NCU application compared to PU can be attributed to Nimin in the neem oil coating, which enhances the population of methanotrophic bacteria, thereby increasing CH_4 oxidation [11,100]. Similarly, the application of KCU resulted in lower CH_4 emissions, which can be attributed to the slower nitrification and higher conservation of NH_4^+-N released from urea. The presence of karanjin inhibits nitrification, thereby increasing soil NH_4^+-N levels as well as enhancing the activity of methanotrophic bacteria, which consume CH_4 more efficiently [101]. Previous reports have shown varying effects of NIs on CH_4 emissions, with some studies indicating a reduction or no effect, while others suggest potential increases due to higher NH_4^+-N conservation leading to a rise in nitrifier populations over methanotrophs [102–105]. However, our findings support the role of neem and karanj oil in mitigating CH_4 emissions. The significantly lower emissions of CH_4 measured in NCU and KCU compared to PU may be attributed to the presence of Nimin in the neem oil coating and karanjin in karanj oil coating which have been reported to increase the population of methanotrophic bacteria in the soil, thereby enhancing CH_4 oxidation and reducing emissions [11,100].

The lowest CH_4 emissions were observed from SCU, which can be attributed to the role of sulphur as an electron sink, scavenging acetate and hydrogen to inhibit methanogenesis. Also, sulfate deposition have been reported to reduce CH_4 emissions from wetland sources [106]. This also aligns with findings by Linquist et al. [107], who reported a 40 %

reduction in methane emissions from paddy fields with ammonium sulfate compared to urea. Additionally, the slow-release properties of SCU and the competition between methanogens and sulfate-reducing bacteria for substrates, contribute to this reduction [108,109].

Methane emission was found to be negatively correlated with dehydrogenase activity (DHA), which is an indicator of soil respiration rates [50] (Fig. 4), while highest DHA was observed in SCU. This was likely due to the enhanced aerobic microbial activity due to S ions and controlled urea release, which resulted in anaerobic conditions favouring CH_4-producing microbes, thus lowering CH_4 emissions effectively [110]. The highest CH_4 emissions were observed from PU, which can be attributed to methanotrophs switching their substrate from CH_4 to NH_4^+ due to the enzymatic similarity, thus inhibiting CH_4 oxidation. Additionally, the nitrogenous fertilizer stimulates plant growth, providing more carbon substrates to methanogens in the rhizosphere [11,111].

The CO_2-equivalent emissions were similar across both years, with CH_4 being the dominant contributor. Notably, the control (Con) plots with no fertilizer showed high CO_2-equivalent emissions, at 545.2 and 528.6 kg CO_2 eq. ha^{-1} for each respective year due to CH_4 emissions. The highest overall emissions was observed with PU, which was equally contributed by CH_4 and N_2O. Except for SCU in 2020, the CO_2-eq was comparable in both the years among NCU, KCU and SCU. Also, no significant difference was observed in the mean CO_2-eq between NIs and SCU. Taking into consideration the economic yield benefits, SCU application resulted in a substantial decrease in GHGi followed by NCU and KCU, while highest GHGi was observed in PU. These reflected the GHG mitigation potential of these treatments while considering economic yield.

4.3. Indicators of soil health: soil nutrient, microbial and enzymatic activities

While indicators such as microbial biomass carbon (MBC) and nitrogen (MBN) are crucial for nutrient cycling and reflect changes in soil conditions [52], GHG emissions are also microbially mediated processes that depend on available nitrogen sources, specifically NO_3^--N and NH_4^+-N [112]. Dehydrogenase activity (DHA) assesses soil's oxidative processes and overall health, while nitrate reductase activity (NRA) and urease activity indicate the efficiency of nitrogen cycling and fertilizer utilization [50,53,54]. Appropriate indicators of soil health should include nutrient, microbial, and enzymatic activities as they comprehensively reflect the soil's capacity to sustain biological productivity, nutrient cycling, and respond to environmental changes [113]. Urea applications significantly enhanced soil NO_3^--N and NH_4^+-N compared to the unfertilized Con, promoting rapid NH_4^+-N and NO_3^--N release that can lead to N loss via volatilization [114]. Coated urea on the other hand, provided a more sustained release of N, throughout the growing season, potentially benefiting plant growth [115]. Post-fertilizer application, NH_4^+-N concentrations peaked rapidly within 2–7 days, followed by a decline as nitrification converted NH_4^+-N to NO_3^--N [116]. The NO_3^--N levels subsequently increased, with peak concentrations occurring approximately one week after application, suggesting a temporal shift from NH_4^+-N dominance to NO_3^--N accumulation [117,118]. However, the release rate of these N forms is not linear, as it is influenced by plant nutrient uptake needs and soil microbial activity [119]. Microbial activity plays a crucial role in this process, with significant implications for N availability and soil health. The use of NIs such as NCU reduce NO_3^--N losses while enhancing soil available N [120]. However, an increase in cumulative NH_3 emissions with NCU compared to PU is observed. This is attributed to the inhibition of nitrification by neem oil coating, resulting in higher concentrations of NH_4^+-N, which can lead to increased NH_3 emissions under alkaline soil conditions [11]. The SCU demonstrated lower N losses compared to PU, likely due to the addition of S to the soil, which supports microbial activity. Soil NH_4^+-N content was positively correlated with the activities of enzymes involved in C, N, and S cycling. This correlation is attributed to high crop biomass and sufficient N supply, which increases substrate availability for microbial processes and enzyme activities [121].

With urea treatments enhanced microbial and enzymatic activities were observed. This could be due to the increase in the soil available nutrients with urea application, which provide C substrates and nutrients for the growth and reproduction of soil microorganisms [122,123]. Microbial activity peaked during the flowering stage of the crop, indicating a tendency of microbes to assimilate more C and N for their energy requirement and maximum nutrient translocation from soil to plant [124–126]. The rhizospheric microbial activity is at its peak during the flowering stage, leading to higher values of soil MBC and MBN [127]. Urea-treated soils also exhibit higher belowground biomass, contributing further to increased MBC and MBN [128]. Comparatively, urea coating enhanced soil MBC but reduced MBN, likely due to slower N release compared to uncoated urea (PU) [129,130]. In addition to delaying the hydrolysis of urea, the addition of coated urea also increased the content of organic carbon, thereby increasing MBC [131]. However, coated urea treatments, particularly KCU, showed significantly higher MBN, possibly due to increased dissolved organic carbon availability promoting microbial growth [132,133].

The highest enzymatic activities during the flowering phase followed by physiological maturity indicated a relationship between soil microbial metabolic activity and plant growth stages [134]. The DHA in soil represents the overall metabolic status and has been widely used as an index of microbial activity [135]. The increase in DHA due to urea application is attributed to the availability of higher C substrates, being the sole sources of C and energy for heterotrophs [136]. Our findings are in line with previous studies which showed a significant increase in DHA in plots treated with fertilizer [112,137]. The application of coated urea treatments exhibited enhanced DHA compared to PU, suggesting a potential enhancement of microbial metabolic processes with coated urea application [138]. The high DHA observed in KCU, can be attributed to the increased availability of N for microorganisms, which enhances soil nitrification activities [139]. On the other hand, SCU had the highest DHA due to its gradual release of S and N, which sustains microbial growth and activity, enhancing overall soil microbial processes [41,140, 141].

Soil NRA which catalyzes the reduction of NO_3^- to NO_2^-, is one of the important enzymes involved in the soil denitrification process [142]. Nitrification inhibitors have the capacity to efficiently impede the NH_3 oxidation process by suppressing the growth of NH_3-oxidizing archaea bacteria resulting in decreased soil NRA [143]. Reduced NRA in paddy soil limits the initial conversion of NO_3^- to NO_2^-, thereby decreasing the substrate availability for subsequent denitrification steps mediated by nitrite reductase, nitric oxide reductase, and nitrous oxide reductase. This reduction in NRA ultimately results in lower N_2O production and emissions as observed in NCU and KCU [144,145]. The SCU, promotes more sustained NRA in the soil due to its gradual nutrient release, which might be due to mechanisms similar to those observed in the NIs, resulting in significantly lower NRA compared to PU. On the other hand, application of PU leads to soil acidification through nitrification, increasing the N_2O:(N_2O + N_2) product ratio. This, coupled with the high NRA observed in PU-applied soil, results in enhanced N_2O emissions under denitrifying conditions [146].

Urease enzyme in soil hydrolyzes urea to CO_2 and NH_3, thus playing a crucial role in N cycling [147]. Urease activity is widely used to assess the impact of soil management practices on soil quality [148]. Also, increased urea substrate concentration stimulates urease activity, which is consistent with our observation in PU [149]. The rapid increase in soil NO_3^- concentrations due to urease-catalyzed urea hydrolysis aggravates N losses through N_2O emissions, as urease activity is highly sensitive to soil NO_3^--N concentrations [150]. Since no significant differences were observed among the coated urea fertilizers, it can be concluded that, compared to PU, coated urea releases N slowly and continuously, reducing urea substrate concentration and soil urease activity, thereby mitigating N loss [151].

Reported SQI values in long-term studies across the Indo-Gangetic Plains typically range from 0.3 to 0.7 [74,152], whereas the values observed in our study (−0.47 to 0.52) were relatively conservative, likely reflecting the shorter experimental duration, where noticeable restoration or enhancement of soil quality is still in early stages. The negative SQI scores for PU in both the years, indicate poorer soil quality and microbial functioning relative to EEFs [153]. The strong positive loadings of MBC and DHA on PC1 suggest that microbial biomass and dehydrogenase activity were the primary drivers of soil quality variation [154]. Conversely, negative PC1 loadings of NRA and urease suggest a distinct functional axis, where rapid enzymatic nitrogen turnover is potentially decoupled from microbial biomass development [155]. The negative positioning of PU indicated higher association with nitrate reductase and urease activity, suggesting rapid nitrogen turnover and potential N losses [156]. In contrast the positive grouping of SCU, KCU and NCU with MBC and DHA signified enhanced microbial function. Among all the EEFs, SCU showed the strongest association with microbial driven soil quality, while KCU and NCU showed moderate enhancement, reflecting stabilizing effects of nitrification inhibition and soil microbial responses [157].

These soil quality trends also closely mirrored agronomic and GHG outcomes. For instance, treatments with higher SQI-PCA scores, particularly KCU and SCU, not only achieved higher yields and nitrogen uptake but also exhibited lower GHG emissions and GHG intensity. In contrast, PU showed the poorest SQI, weakest crop performance in terms of yield and nitrogen uptake, and the highest GHG emission despite receiving equivalent nitrogen inputs. These results underscore that the use of EEFs through coating and controlled release mechanisms which can significantly improve soil health, increase nitrogen recovery, and reduce N losses compared to conventional uncoated urea.

4.4. Enhanced efficiency fertilizers and its greenhouse gas intensity mitigation potential

The use of NCU has been made mandatory in India, effectively replacing PU as a source of N fertilizer due to its beneficial nitrification inhibition properties and ability to enhance NUE of crop [11,81]. Use of NCU is also promoted by the government and scientists due to its ability to promote beneficial microbial growth and improve soil enzymatic activities [158]. Our findings suggest significant reduction in GHG emissions and GHGi on application of NI (NCU and KCU) and SRF (SCU) under the respective scenarios. The use of higher concentration of KCU showed promising results in GHG mitigation and were at par with NCU. On the other hand, SCU showed highest potential in reducing GHGi from rice cultivation under the Rice-wheat cropping system (RWCS) in Indo-Gangetic Plains region. In addition to reducing the gaseous emissions from rice cultivation these EEFs have showed remarkable potential in improving soil health by stimulating microbial biomass and enzymatic activities. Given that rice-wheat cropping systems dominate large parts of South Aisa and China [159], similar trails and projections across these agro-ecologies would help validate the broader applicability and efficacy of the EEFs in mitigating GHG emissions. Also, in light of India's commitment to the Paris Climate Change Agreement through its nationally determined contributions [160], these EEFs could contribute to reducing India's contribution to global agricultural emissions.

4.5. Economic implications of coated fertilizer treatments

The results indicated that both KCU and SCU application not only increased crop productivity and reduced GHG emissions but also performed better than PU. After deducting the total cost of cultivation from gross return, the net return of KCU and SCU even outweighed NCU by 3746 R ha^{-1} and 4399 R ha^{-1} (45–52 USD ha^{-1}), respectively. Notably, the net benefit of using alternatives like KCU and SCU is due to their positive impact on increasing the grain and straw yield which negated the coating cost [161,162] Not only coating urea with NIs and SRFs offer economic benefits, but further gains can be achieved by adjusting application rate and timing [163]. Integrating these fertilizers with other management practices such as irrigation, tillage, and straw retention can also enhance their economic effectiveness [164]. The net benefit of using these EEFs would be even greater if the environmental cost of reduced GHGi is also considered.

5. Conclusion

The findings of this study suggest that EEF, including sulphur-coated urea (SCU), and karanj-coated urea (KCU) have the potential to reduce both CH_4 and N_2O emissions and improve soil microbial biomass and enzymatic activities in rice cultivation systems. Projections based on the results indicate that application of NCU in the IGP region during rice cultivation reduces the GHGi of rice by 12.2 %, while adopting SCU may achieve a reduction of 25.8 % in GHGi, along with significant improvement in the soil health indicators highlighting their remarkable mitigation potential. These benefits were also corelated with improved soil quality and economic returns, especially under SCU and KCU, supporting their role in sustainable and profitable rice cultivation. However, with the study's limited scope further long-term research is needed to validate the findings across different agroecological contexts and over time. In summary, adopting EEFs, particularly SCU and alternatives like KCU, show promise as a strategy to reduce GHG emissions and enhance soil health in rice cultivation systems, with the potential to support India's GHG reduction targets under the Paris Climate Change Agreement.

CRediT authorship contribution statement

Ankita Paul: Writing – original draft, Software, Investigation, Formal analysis, Data curation. **Arti Bhatia:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Conceptualization. **Julia Drewer:** Writing – review & editing, Project administration, Funding acquisition. **Ritu Tomer:** Data curation. **Vinod Kumar:** Resources, Formal analysis. **Shikha Sharma:** Formal analysis. **Namita Das Saha:** Supervision, Methodology. **Bidisha Chakrabarti:** Supervision, Methodology. **Y.S. Shivay:** Resources, Methodology. **Robert M. Rees:** Writing – review & editing, Project administration, Funding acquisition. **Mark A. Sutton:** Writing – review & editing, Project administration, Funding acquisition.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jafr.2025.102376.

Data availability

Data will be made available on request.

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