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Assessment of carbon carrying capacities of Alfisol and Vertisol under long-term manuring and fertilization

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Running head: Assessment of maximum C carrying capacity

Highlights

- The maximum carbon (C) carrying capacities of Alfisols and Vertisols were estimated.
- Higher carbon saturation deficit marked higher carbon sequestration.
- Application of 100%NPK + lime/FYM showed highest build-up of carbon in soil.
- Carbon input dosage (including FYM) should be soil C saturation level based.
- Complex interaction of C input, clay minerology, texture effects soil C stability.

ABSTRACT

Carbon (C) sequestration in soil is limited by the soil's maximum C carrying capacity (C_m) and C saturation deficit (S_d). These two parameters have enormous implication for choosing appropriate management strategies to increase a soil's C sequestration potential (CSP). The present study aims to estimate the C_m and S_d of two Alfisols from Bhubaneswar (Alfisol-BHNS) and Pattambi

(Alfisol-PTMB) and two Vertisols from Parbhani (Vertisol-PRBN) and Junagadh (Vertisol-JNGD) in India with a history of long-term manuring and fertilization practices. Both the Alfisols maintained a rice-rice cropping system whereas the Vertisols had a soybean-safflower and groundnut-wheat cropping system, respectively. Irrespective of soil types and cropping systems, the soil organic C (SOC) content and stock were the highest in the treatment receiving 100% of recommended dose of NPK combined with 10 Mg ha^{-1} well decomposed farmyard manure (100% NPK + FYM). The mean annual estimated return of C (MAERC) across the soil types followed in the order of 100% NPK + FYM> 150% NPK = 100% NPK + Lime (Agricultural grade, purity 80%)> 100% NPK > 50% NPK > control. The C_m at 0-30 cm soil depth was 39.3, 21.7, 47.6 and 25.0 Mg ha^{-1}, respectively for Alfisol-PTMB, Alfisol-BHNS, Vertisol-JNGD and Vertisol-PRBN. The S_d value was the highest in the control treatment and lowest in100% NPK + FYM treatment, signifying a higher CSP of the former soil than the later. Therefore, 100% NPK + FYM treatment would require a minimum threshold level of C addition to maintain the SOC at equilibrium. The estimated minimum threshold limit of annual C input (A_E) to preserve the existing SOC level was 1.84, 0.39, 1.57 and 0.90 Mg ha^{-1} yr^{-1} for Alfisol-PTMB, Alfisol-BHNS, Vertisol-JNGD and Vertisol-PRBN, respectively. Alfisol-PTMB and Vertisol-JNGD showed a higher CSP than Alfisol-BHNS and Vertisol- JNGD because the former two soils had higher S_d values than the later soils. The MAERC, climate, soil texture and clay mineralogy largely influenced the SOC sequestration and C_m values. An integrated application of 100%NPK with either lime or FYM is recommended to maintain the sequestration and stabilization of SOC in the studied soils. The CSP of the soils could be further increased via innovative approaches such as increasing soil's reactive mineral contents to lock additional C.

Keywords: Alfisol, Vertisol, C saturation deficit, Decay and humification rate constants, Maximum C carrying capacity, SOC stabilization

1. Introduction

Soil organic carbon (SOC) is a key soil property considered as backbone of soil quality (Ghosh et al., 2021) and plays a crucial role in greenhouse gas balance and food production (Lorenz and Lal, 2016). Soil management practices such as excessive tillage and sub-optimal and imbalanced fertilizer application, and limited organic recycling have contributed to low SOC content (less than 10 g kg^{-1}) in most of the soils of India (Nayak et al., 2012; Lal, 2004; Rudrappa et al., 2006). The balance between carbon inputs (manures and crop residues) and outputs (decomposition) governs the dynamics of SOC (Bai et al., 2019). The amount of SOC can be enhanced by capturing and securing atmospheric CO_2 into the soil as SOC by the process known as sequestration. SOC sequestration plays an important role in restoring soil quality, creating climate smart agro-ecosystems, and enhancing input use efficiency. With the intention to foster food security and decrease CO_2 concentration in the atmosphere, the 4 per 1000 initiative was proposed at the UN Framework Convention on Climate change (UNFCC, COP21) to increase SOC stocks by 0.4% per annum (Chen et al., 2019). Consequently, the feasibility, duration and degree of SOC sequestration in various soil types worldwide have gained immense significance (Minasny et al., 2017; van Groenigen et al., 2017). The depth of the soil, clay content, mineralogy, nutrient reserve and initial SOC stock are some of the key factors that determine the SOC sequestration capacity (Lal, 2018). The maximum attainable SOC level in soil is critical in determining C sequestration potential (CSP) of a soil. There is an upper limit to SOC storage and soil C saturation level, which is not dependent on land use or climate, and is not subjected to decomposition due to mineral

protection (Schmidt Rohr et al., 2004, Stewart et al., 2007, Guillaume et al., 2022). Soil C saturation is the level of maximum amount of C that can be sequestered physically and chemically, and this level is likely to be greater than the current level of sequestration. The soil C saturation deficit (S_d) will decrease as the soil approaches C saturation. Therefore, before designing the C input rate, it is essential to know the degree of C saturation level of a specific soil. The C saturation level of soils depends on different soil types with different C input, texture, clay mineralogy and climate (Stewart et al., 2007, 2008).

Enrichment of C in soil is the result of positive balance between humification and decay of added C substrates, and these processes are impacted by various biotic and abiotic factors (Purakayastha et al., 2008). Use of locally available organic amendments such as farmyard manure (FYM), crop residue, green manure and biofertilizer play an important role in C sequestration (Mahajan and Gupta, 2009; Ghosh et al., 2010, Purakayastha et al., 2019). Among soil factors, texture plays an important role in C sequestration. More mineralization of C and N was recorded in coarse-textured soils compared to fine-textured soils (Hassink, 1992), which could be due to the greater ability of fine-textured soils to physically protect SOC against microbial attack (Hassink, 1995). Chemical adsorption of C onto the surface of clays and clay minerals and physical occlusion within microaggregates (Sissoko and Kpomblekou-A, 2010; Singh et al., 2018) make SOC inaccessible to microbial degradation in clay-dominated soils. Once the mineral fraction of soil is saturated, additional SOC accumulation occurs at the poorly protected coarse fraction of soil, which results in a rapid turnover rate (Six et al., 2002; Guillaume et al., 2022). Type of phyllosilicates also influences SOC stabilization (Chevallier et al., 2003; Singh et al., 2018). The decomposition of organic C (OC) in soil by heterotrophic microorganisms by different phyllosilicates followed the order: montmorillonite>hectorite>illite>kaolinite, which is inversely related to the binding

capacity of the phyllosilicates (O'Loughlin et al., 2000). However, the mean residence time of decomposed OC is estimated to be remarkably higher for smectite- dominated soil. Alfisols are strongly weathered base cation rich soils dominated by 1:1 type clay minerals (e.g., kaolinite), while Vertisols are characterized by higher content of 2:1 type clay minerals (e.g., smectite, montmorillonite) with swell-shrink properties. These two soil types therefore would have different SOC stabilization properties leading to various degrees of C sequestration.

Long-term fertilizer experiments in India are continuing in various agro-climatic regions under contrasting soil types which could be unique study sites for assessment of C S_d. Integrated application of 100% recommended dose of NPK with well decomposed FYM is a popular management practice among Indian farmers (Purakayastha et al., 2008; Meena et al., 2019; Shahid et al., 2016) because this practice has proved enhanced soil fertility and productivity across the country (Majumder et al., 2008; Datta et al., 2018). An additional benefit of the above nutrient management practice is SOC sequestration, which has been viewed as a climate change mitigation option. However, a major problem of the integrated application of 100% NPK with FYM is: FYM is applied as a blanket recommendation without considering the C saturation level of soils. A blanket recommendation of FYM at the rate of 5 Mg ha^{-1} (Mandal et al., 2007), 10 Mg ha^{-1} (Srinivasarao et al., 2012a; Datta et al., 2018) and 15 Mg ha^{-1} are applied in different Indian cropping systems (Srinivasarao et al., 2012a; Purakayastha et al., 2008; Rudrappa et al., 2006) and elsewhere (Liang et al., 2019). In view of limited availability of organic resources e.g., FYM in rural India, it is essential to apply these materials precisely depending upon the C saturation deficit of soil.

To date, no study has been carried out in India to determine the C saturation level of Alfisols and Vertisols located in different agro-climatic regions that support the cultivation India's major

144 agricultural crops and represent soils with distinguishable texture and mineralogical makeup.
145 Enrichment and stabilization of C is also greatly influenced by the chemistry of C inputs (i.e., C:N
146 ratio and lignin, polyphenol and initial N contents) and soil microbial community structure that
147 trigger C cycling and humification process (Baumann *et al*., 2009) in different soil types.
148 Information on C carrying capacities of major soil orders located in various agro-climatic regions
149 would help in designing efficient nutrient and manure management strategies for long-term C
150 sequestration, which is presently lacking in India and many other countries. The saturation deficit
151 of soil C estimated from the soil's maximum C carrying capacity and present level of C as
152 envisaged from the current study would help assessing of CSP in agro-ecological region basis. The
153 soil thus reaching saturation needs lower C inputs than that is away from the C saturation point.
154 Thus, the information on soil's C saturation level is essential to save precious carbonaceous
155 material and reduce emissions of CO_2.

156 The present study hypothesized that similar/dissimilar soil orders might have differential C
157 carrying capacities triggered by differences in soil texture, clay mineralogy, crop and nutrient
158 management practices and climate. We also hypothesized that some nutrient management
159 treatments such as 100% NPK, 100% NPK+ Lime and 100% NPK+FYM might have
160 approached/reached C saturation levels in the studied Alfisols and Vertisols located in different
161 agro-climatic regions.

162 In this study, we investigated the following questions:

163 1. What was the mean annual estimated return of C (MAERC) in various manuring and
164 fertilization practices in different cropping systems under Alfisol and Vertisol?
165 2. What are the dominant factors that control the C sequestration and maximum C carrying
166 capacities of these soils?

3. Do the decay and humification of added C differ in all the four soils leading to different SOC sequestration and stabilization?

4. What is the threshold limit of C input required to maintain SOC at equilibrium and maximum C carrying capacities of spatially separated Alfisols and Vertisols?

To address the above questions, we assessed the mean annual estimated return of C (MAERC), decay rate constant (k), humification rate constant (h), minimum C input required to maintain SOC at equilibrium (A_E), maximum C carrying capacities (C_m) and saturation deficits (S_d) of two each of Alfisol and Vertisol soils under long-term manuring and fertilization trials located in different agro-climatic zones of India.

2.Materials and methods

2.1. Experimental sites and soil types

The study was conducted at four different ongoing long-term fertilizer experimental (LTFE) sites in India under All India Coordinated Research Project on Long Term Fertilizer Experiments (LTFE) of the Indian Council of Agricultural Research (ICAR). Three distinct cropping systems at the above sites were investigated as the following: rice (*Oryza sativa* L.)−rice at Pattambi in Kerala and Bhubaneswar in Odisha, groundnut (*Arachis hypogeae* L.)−wheat (*Trticum aestivum* L.) at Junagadh in Gujarat, and soybean (*Glycine max* L.)–safflower (*Carthamus tinctorius* L.) at Parbhani in Maharashtra. All the above LTFEs were started in 1996 except at Bhubaneswar which was started in 2002. The site details including their climate, temperature, rainfall, cropping systems, and manuring and fertilization details are provided in Supplementary Information (Table S1). The climate of Pattambi, Bhubaneswar, Junagadh and Parbhani is tropical and sub-humid,

per-humid, dry sub-humid and semi-arid, respectively. The average annual rainfall of these locations is 2727, 1449, 874 and 844 mm, and the average temperature is 32, 33, 28.3 and 26.1°C, respectively.

The classification, mechanical and physico-chemical properties of the experimental soils are given in supplementary Table S2. The soil of Pattambi (Alfisol), Bhubaneswar (Alfisol), Junagadh (Vertisol) and Parbhani (Vertisol) is classified as Typic Haplustalf, Aeric Haplustalf, Vertic Ustochrept and Typic Chromusterts, respectively, according to the United States Department of Agriculture (USDA) classification system. The soils of Pattambi (pH =5.5) and Bhubaneswar (pH = 6.4) were acidic in reaction, while that of Junagadh (pH = 8.5) and Parbhani (pH = 7.7) were alkaline in reaction. The soils of Pattambi and Bhubaneswar were sandy clay loam and sandy loam in texture, whereas soils of Junagadh and Parbhani were clayey in texture. The soils at Pattambi and Bhubaneswar had SOC of 15 and 7 g kg^{-1} and that at Jungadh and Parbhani was 16 and 55 g kg^{-1}, respectively. The SOC contents in Pattambi and Junagadh were high whereas in Bhubaneswar and Parbhani these were low.

2.2. Cropping systems and management practices

The treatments considered for the present investigation under LTFE trials comprised of control (no fertilizer or manure), 50% nitrogen-phosphorus-potassium (NPK) of the recommended dose, 100%NPK, 150% NPK, 100%NPK+ farmyard manure (FYM) and 100%NPK + Lime. The recommended fertilizer dose of different crops at Bhubaneswar, Pattambi, Junagadh and Parbhani sites are given in supplementary Table S3. The nutrients N, P and K were applied at all sites as urea, diammonium phosphate and potassium chloride, respectively. Agricultural grade lime with

85% purity was applied at the rate of 0.5 Mg ha^{-1} to rice crop at Pattambi and Bhubaneswar sites only. Lime was not applied at Junagadh and Parbhani sites because these soils were alkaline in nature. Farmyard manure (FYM), with an average composition of 250 g kg^{-1} C, 5 g kg^{-1} N, 2.5 g kg^{-1} P and 15 g kg^{-1} K, was applied at a rate of 5 Mg ha^{-1} in each crop season, i.e., twice in a year, for all the cropping systems at all four locations. Before ploughing in the wet season (July–November) and winter season (November-April) rice crop, well-decomposed FYM was applied at the rate of 5Mg ha^{-1} per season to the specified plots of the experiment and mixed well with the soil. The plots were pre flooded for one week followed by puddling and leveled one week before transplanting of rice seedlings (30 and 40 days old for wet and dry seasons, respectively) at a spacing of 0.20m × 0.20m in July (for wet season) and 0.20m × 0.15m in January (for winter season). The size of the plot was 15 m x 10 m (150 m^2) and 12.5 m x 10 m (125 m^2) at Bhubaneswar and Pattambi, respectively. The experiment was laid out in 4 replications with spacing of 1.2 m between the plots. 72 soil samples were collected from each soil depth (0-15 cm and 15-30 cm) from each experimental site under Alfisols.

The plot size was 15 m x 10 m (150 m^2) and 13.5 m x 10 m (135 m^2) at Junagadh and Parbhani, respectively. The experiment was laid out in randomized block design with 4 replicates at a spacing of 1m between the plots at both the experimental sites.

2.3. Collection of soil samples and assessment of soil carbon

The soil samples were collected to a depth of 0-15 and 15-30 cm randomly from three sampling spots in each plot with a soil core sampler, removed from the core, mixed and composited. The soil sampling depth up to 30 cm was chosen as majority of the root biomass of

233 the stated field crops remained concentrated within 30 cm soil depth (Soil Sampling Techniques
234 Advanced Soil Fertility, 2006).The soil samples were collected after harvest of rice, groundnut
235 and soybean in the year 2015, Total 48 (6 treatments x 4 replications x 2 soil depths) and 40 (5
236 treatments x 4 replications x 2 soil depths) composite soil samples each representing Vertisol and
237 Alfisol, respectively were prepared. Three sets of soil core collected for estimation of bulk density
238 (BD) the date of which is presented in Table S4. The total soil carbon (TSC) was determined by
239 dry combustion by packing samples in silver capsules and further analyzed in a CHNS analyzer
240 (Euro EA 3000, Eurovector). The soil organic C (SOC) was determined by the difference between
241 TSC and soil inorganic C (SIC), after removing inorganic C by treating with 2N H_2SO_4. The depth-
242 wise data of SOC is presented in supplementary Table 2. The SOC stock was computed for
243 individual soil depth as follows:

244 SOC stock (Mg ha^{-1}) = A × B × C × D Eq. 1

245 where, A= area (1 ha = 10000 m^2), B= Bulk density (Mg m^{-3}), C= C content in % /100, and D=
246 soil depth (m).

247 The C stock for 0-15 and 15-30 cm soil depth was summed up to get the C stock in 0-30 cm soil
248 depth.

249

250 *2.4. Assessment of mean annual estimated return of carbon (MAERC)*

251 The mean annual estimated return of carbon (MAERC) input was calculated on the basis of
252 observed and derived values of C content in root, rhizodeposition, stubble biomass and leaf fall of
253 various crops in the cropping systems. The contributions of C from various sources are provided
254 in supplementary Table S5. The root biomass was estimated by the established aboveground

biomass: root ratio of the respective crops in the cropping systems. The total root biomass yield of rice/wheat was computed as 27% of the total above ground biomass harvested at maturity (Bronson et al., 1998). The stubble biomass was measured as 12% of total straw biomass for both rice and wheat. The C content of root biomass of rice and wheat was estimated to be 32% and 34%, respectively. The contribution of C through rhizodeposition from rice was estimated as 85% of total root biomass C and 1.4 times root C in wheat (Das et al., 2013). For groundnut, the root biomass was estimated as 40% above ground biomass, nodule biomass as 11.3% of root biomass, rhizodeposition C as 10% of above ground biomass and the leaf fall and stubble remaining as 12% of above ground biomass yield (Srinivasarao et al., 2012b). The C content in the leaf, root and nodules of groundnut was estimated as 35.5, 34.4 and 35.6 %, respectively (Srinivasarao et al., 2012b). The root biomass of soybean was computed as 38.5% of above ground biomass, leaf fall 15% of haulm yield, nodule biomass as 9.3% of above ground biomass, stubble biomass 5 % of above ground biomass and rhizodeposition C as 10% of above ground biomass (Bartholomew et al.,1966; Shamoot et al., 1968; Srinivasarao et al., 2012a). The C content in the leaves, stubble, roots /nodules of soybean was estimated as 37.8, 34.2 and 31.5 %, respectively (Srinivasarao et al., 2012a). In safflower, leaf fall biomass, stubble biomass and root biomass were 12, 5 and 37.8%, respectively of above ground biomass, and rhizodeposition C was 1.4 times of root biomass C (Srinivasarao et al., 2012a). The C content in the leaves, stubbles and roots of safflower were estimated as 35.5, 32 and 34.4 %, respectively (Srinivasarao et al., 2012a). The cumulative C input (stubble, root and rhizodeposition) into soil was calculated. The C input added externally into soil through FYM was also estimated with average C content of 35%. Following the above procedure, detailed estimate for crop derived/residue C returned to the soil, along with external addition for individual crop in each cropping system was calculated and these were summed up to ascertain

278 total estimated return of C and dividing these values by number of years of the experiment yielded
279 MAERC.

280 Sequestration and stabilization of SOC were calculated by using the following formulae:

281 SOC sequestration (Mg ha^{-1}) = (SOC_{trt} — $SOC_{initial}$). Eq. 2

282 SOC stabilization (%) = (SOC_{trt} — $SOC_{initial}$)/MAERC Eq. 3

283 SOC_{trt}, $SOC_{control}$, $SOC_{initial}$ indicate soil organic C stock in 0-30 cm soil depth in different fertilized
284 treatments, control (no fertilization) and initial (before start of trial) in the same soil depth,
285 respectively. The $SOC_{initial}$ stocks in 0-30 cm soil depth were calculated as 24.75, 12.1, 30.1, and
286 14.0 Mg ha^{-1} by using the following formula in Alfisol (Pattambi), Alfisol (Bhubaneswar), Vertisol
287 (Junagadh), and Vertisol (Parbhani), respectively.

288 SOC stock (Mg ha^{-1}) = A x B x C x D Eq. 4

289 A= Area of 1 ha is 10000 m^2

290 B = Bulk density in Mg m^{-3}

291 C = SOC content in %/100

292 D= Soil depth in m

293 The SOC stock in 0-15 and 15-30 cm were calculated separately and then these values were
294 summed to get SOC stock at 0-30 cm soil depth.

295

296 *2.5. Assessment of maximum carbon carrying capacity and carbon saturation deficit (S_d)*

297 Carbon saturation model was used to estimate C saturation deficit (S_d) and maximum C carrying
298 capacity (C_m). The C saturation model has a whole soil saturation limit due to inherent
299 physicochemical limitations with the following expression (Stewart et al., 2007):

300 $dC_t/dt = I\ (1- C_t/C_m) – kC_t$ Eq. 4

301 where, C_m is the maximum amount of C that can be stabilized by the soil. The C_t denotes SOC
302 content in soil. In this model, soil C storage is limited by S_d which is defined as:

303 $S_d= 1- C_t/C_m$ Eq. 5

304 Decay constant, "*k*" of soil belonging to a particular site was estimated by plotting the data of
305 change in SOC over year (dC_s/dt) versus mean annual estimated return of C (MAERC) inputs data
306 and equating the relationship with $dC_s/dt=hA – kC_s$ (y=mx+C) (Jenkinson, 1988), where slope
307 represents humification rate constant, "*h*" of the straight line curve and intercept represents "kC_s".
308 The value of "*k*" was substituted in the above equation and the maximum C carrying capacity,
309 "C_m" and saturation deficit, "S_d" were estimated where C_s is the initial SOC in pool S, and A is
310 the MAERC input data during the period of study. We assumed that whole amount of C is derived
311 from stubbles, roots, nodules and rhizodeposition remaining in the 0-30 cm soil layer. The amount
312 of C input required to maintain the SOC at equilibrium (A_E) was estimated by making dC_s/dt equal
313 to 0 and $A=kC_s/h$.

314

315 *2.6. Comparative assessment of carbon saturation by incubation experiment*

316 An incubation study was conducted to compare and correlate the S_d computed from the carbon
317 saturation model with that derived from the substrate C driven saturation experiment. The C
318 saturation level of individual treatment was achieved by incubating the 35 g soil taken in a 50 ml

capacity beaker with varying doses of C (starch as the C source) at the rate of 0.5, 1.0, 2.0 g C kg^{-1} soil, sealed with parafilm for preventing moisture loss and proper aeration, moisture maintained at 2/3rd water holding capacity for 15 months in a BOD incubator at 28°C and the SOC content was estimated in a CHNS analyzer (Euro EA 3000, Eurovector) at the end of incubation. The data pertaining to SOC content (x-axis) and varying doses of added substrate C (y-axis) in all the long-term fertilization treatments was fitted in straight line relationships (y = mx + C, m is the slope, and C is the intercept) in window-based MS-Excel program. The slope of fitted line (incubation experiment) was noted to understand its association with C S_d as estimated from C saturation model (Stewart et al., 2007).

2.7. Statistical analysis

The data pertaining to SOC stock, SOC sequestration and SOC stabilization were analyzed in a two-factor randomized block design (RBD) using SAS program version 16.0. We used the same program for the analysis of variance (ANOVA) and the mean data were statistically separated by Duncan Multiple Range Test at P=0.05. The mean annual estimated return of carbon data was analyzed on one way ANOVA in RBD for a particular experimental site. The relationship between dC_s/dt versus mean annual estimated return of C (MAERC) was performed in Microsoft EXCEL program for estimation of decay rate constant, '*k*', humification rate constant, '*h*' and annual C input required to maintain SOC at equilibrium (A_E). The correlation coefficients (r) between C S_d with change in SOC per unit change in substrate C (slope) in the incubation experiment was estimated in SAS.

3. Results

3.1. Mean annual estimated return of carbon (MAERC)

3.1.1 Alfisols

The mean annual estimated return of C (MAERC) was calculated over the years from various sources in wet season and winter season rice under Alfisols from Pattambi (Alfisol-PTMB) and Bhubaneswar (Alfisol-BHNS) varied between the treatments (Table 1). Among the plant derived C input, the highest contribution was received by root biomass C followed by rhizodeposition C, and stubble biomass C. However, the organic C input from FYM was recorded highest. The MAERC was higher in winter rice than wet season rice in both the Alfisols. The MAERC in Alfisol-PTMB was significantly higher in 100%NPK+FYM (6.10 Mg ha^{-1} yr^{-1}) followed by 100%NPK+Lime (3.40 Mg ha^{-1} yr^{-1}), 150%NPK (2.96 Mg ha^{-1} yr^{-1}), 100%NPK (2.85 Mg ha^{-1} yr^{-1}), 50%NPK (2.61 Mg ha^{-1} yr^{-1}) and control (1.87 Mgha^{-1} yr^{-1}). In Alfisol from Bhubaneswar, the trend was almost the same except 150%NPK (1.92 Mg ha^{-1} yr^{-1}) which contributed more MAERC than 100%NPK+Lime (1.79 Mg ha^{-1} yr^{-1}) (Table 1). Irrespective of the treatments, Alfisol-PTMB showed higher MAERC than Alfisol-BHNS.

3.1.2 Vertisols

The MAERC in Vertisols from Junagadh (Vertisol-JNGD) (6.65 Mg ha^{-1} yr^{-1}) and Parbhani (Vertisol-PRBN) (6.01Mgha^{-1} yr^{-1}) was significantly higher in 100%NPK+FYM (Table 1). In Vertisol-JNGD, the MAERC in control, 50% NPK, 100% NPK, 150% NPK was 2.25, 2.55, 2.92 and 3.04, respectively. In Vertisol-PRBN, the MAERC in control, 50% NPK, 100% NPK, 150%

NPK was 1.55, 2.24, 2.54 and 2.80 respectively. The optimal and super optimal doses of NPK showed higher MAERC than the suboptimal dose of NPK.

Table 1

Mean annual estimated return of C (MAERC) to Alfisols and Vertisols under different cropping systems

Fertilization/Soil	MAERC (Mg ha^{-1} yr^{-1})			
	Alfisols		Vertisols	
	Rice-Rice (Pattambi)	Rice-Rice (Bhubaneswar)	Groundnut-Wheat (Junagadh)	Soybean-Safflower (Parbhani)
50% NPK	2.61d§	1.62e	2.55d	2.24d
100% NPK	2.85c	1.71d	2.92c	2.54e
150% NPK	2.96c	1.92b	3.04b	2.80b
100% NPK + FYM	6.10a	5.45a	6.65a	6.01a
100% NPK + Lime	3.40b	1.79c	-	-
Control	1.87e	0.90f	2.25e	1.55e

§The values followed by different lowercase letters in a particular soil are significant according to Duncan's Multiple Range Test at 5% level of significance

3.2. Stock, sequestration and stabilization of soil organic carbon

The data pertaining to depth-wise distribution of SOC and bulk density of all the four soils are provided in Table 2 and Table S4, respectively, which were used to calculate the SOC stock by using the mathematical expression provided in the materials and methods section.

The SOC in the Alfisol from Pattambi and Bhubaneswar was significantly higher in 100% NPK+ FYM at both the depths (Table 2). This was followed by 100% NPK + Lime, 150% NPK, 100 %

NPK, 50% NPK and control at both the sites. In the Vertisols of Junagadh and Parbhani, the SOC content was significantly higher in 100% NPK + FYM at both depths, followed by 150% NPK in the surface layer. This was followed by 100% NPK, 50% NPK and control at the surface layers at both Vertisols. However, in the subsurface layer, no significant difference was observed between 150% NPK and 100% NPK and between 50% NPK and control at Junagadh. At Parbhani, the SOC in all remaining treatments were at par with each other. Overall, the Alfisol from Pattambi showed higher SOC than that from Bhubaneswar while the Vertisol from Junagadh showed higher SOC than that from Parbhani.

Table 2

Soil organic carbon (SOC) contents in 0-15 and 15-30 cm depths

Treatments	Alfisol				Vertisol			
	Pattambi		Bhubaneswar		Junagadh		Parbhani	
	SOC (g kg^{-1})							
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
50% NPK	9.57e§	5.37c	4.93d	2.09c	10.4c	7.48c	8.09c	5.72b
100% NPK	10.7d	5.50c	5.61c	2.38b	10.5c	9.02b	8.43c	5.51b
150%NPK	11.1c	5.95b	5.78c	2.55b	11.9b	9.72b	9.41b	6.12b
100%NPK+FYM	13.6a	8.10a	8.95a	3.71a	13.5a	11.1a	12.34a	8.73a
100%NPK+Lime	11.7b	5.80b	6.22b	2.47b	-	-	-	-
Control	7.81f	4.68d	4.08e	1.59d	9.49d	7.45c	7.53d	5.47b

§The values followed by different lowercase letters in a particular soil are significant according to Duncan's Multiple Range Test at 5% level of significance

3.2.1 Alfisols

In Alfisols, the mean effect of nutrient management treatment showed that the soil organic carbon (SOC) stock was significantly highest in 100%NPK+FYM followed by 100%NPK+Lime, 150%NPK, 100%NPK, 50%NPK and it was lowest in control (Table 3). Irrespective of the treatments, the SOC stock in Alfisol from Pattambi (Alfisol-PTMB) was 50%higher than Alfisol from Bhubaneswar (Alfisol-BHNS). As compared to control, the increase in SOC stock across all other treatments varied between 9 and 52% in Alfisol-PTMB, while it varied between 15to 95 % in Alfisol-BHNS. The SOC sequestration was highest in 100%NPK+FYM and this treatment showed 14.5 Mg ha^{-1} and 11.9 Mg ha^{-1} in Alfisol-PTMB and Alfisol-BHNS, respectively. On an average the amount of SOC sequestered was 6.55 Mg ha^{-1} in Alfisol-PTMB, while it was 4.62 Mg ha^{-1} in Alfisol-BHNS. The SOC stabilization was highest in 100%NPK+Lime (14 %). In Alfisol-PTMB, the 150%NPK (10.8%), 100%NPK+FYM (12.3%) and 100%NPK+Lime (12.1%) being at par with each other showed significantly higher SOC stabilization than the other treatments, while in Alfisol-BHNS, the SOC stabilization was significantly higher in 100%NPK+Lime than in the rest of the treatments. On the average Alfisol-BHNS showed higher SOC stabilization than the Alfisol-PTMB.

Table 3

Soil organic C (SOC) stock, SOC sequestration and SOC stabilization in two Alfisols

Fertilization/ Soil	SOC stock (Mg ha^{-1})		Mean	SOC sequestration (Mg ha^{-1})		Mean	SOC stabilization (%)		Mean
	Alfisol-PTMB	Alfisol-BHNS		Alfisol-PTMB	Alfisol-BHNS		Alfisol-PTMB	Alfisol-BHNS	
50% NPK	28.1e§	14.2j	21.2E	3.35ef	2.10f	2.73E	4.98e	7.41d	6.20D
100% NPK	30.1d	15.9ij	23.0D	5.35d	3.80ef	4.58D	8.42d	12.3bc	10.4C
150% NPK	31.4c	16.7hi	24.1C	6.65c	4.60de	5.63C	10.8cd	13.5b	12.2B

100% NPK+FYM	39.2a	24.0g	31.6A	14.5a	11.9b	13.2A	12.3bc	12.1bc	12.2B
100% NPK+Lime	33.2b	17.2h	25.2B	8.45b	5.10d	678B	12.1bc	15.6a	14.0A
Control	25.8f	12.3k	19.1F	1.05f	0.20g	0.63F	0.21f	1.11f	0.70E
Mean	31.3A	16.7B		6.55A	4.62B		9.77B	12.2A	

§The values followed by different lowercase (interaction of F × S) and upper case (Main effect of F or S) letters in a column/row are significant according to Duncan's Multiple Range Test at P=0.05, F- Fertilization, S- Soil, FYM- Farmyard manure, PTMB-Pattambi, BHNS-Bhubaneswar.

3.2.2 Vertisols

The SOC was significantly influenced by various nutrient management as well as soil type. On the average the SOC stock was highest in 100%NPK+FYM followed by 150%NPK, 100%NPK, 50%NPK and control (Table 4). As compared to control, the SOC stock in Vertisol-JNGD increased by 45%, 28%, 15%, 6% in 100%NPK+FYM, 150%NPK, 100%NPK and 50%NPK, respectively. On the other hand, in Vertisol-PRBN, the extent of increases was60, 21, 11 and 7% in the same sequence as recorded for Vertisol-JNGD. Overall, the SOC stock in Vertisol-JNGD was almost double that of Vertisol-PRBN. On the average, the SOC sequestration in Vertisol was highest in 100%NPK+FYM followed by 150%NPK and 100%NPK. Vertisol-JNGD showed significantly higher amount of SOC sequestration in 100%NPK+FYM and 150%NPK than that in Vertisol-PRBN. Overall, Vertisol-JNGD showed higher SOC sequestration than Vertisol-PRBN. The SOC stabilization In Vertisols was highest in 150% NPK and 100%NPK+FYM with 12%. No significant difference was observed in the SOC stabilized between Vertisol-JNGD and Vertisol-PRBN.

Table 4

Soil organic C (SOC) stock, SOC sequestration and SOC stabilization in two Vertisols

Fertilization/ Soil	SOC stock (Mg ha^{-1})		Mean	SOC sequestration (Mg ha^{-1})		Mean	SOC stabilization (%)		Mean
	Vertisol-JNGD	Vertisol-PRBN		Vertisol-JNGD	Vertisol-PRBN		Vertisol-JNGD	Vertisol-PRBN	
50% NPK	32.0d§	16.1g	24.0D	1.90f	2.10f	2.00D	4.07f	5.32f	4.70C
100% NPK	34.9c	16.7fg	25.8C	4.80e	2.70e	3.75C	9.07cd	5.83ef	7.45B
150% NPK	38.7b	18.2f	28.5B	8.60d	4.20c	6.40B	15.8a	8.33de	12.0A
100% NPK+FYM	43.9a	24.1e	34.0A	13.8b	10.1a	12.0A	11.6bc	12.7b	12.1A
Control	30.3d	15.1g	22.7E	0.20f	1.10f	0.65D	0.57g	3.84f	2.20D
Mean	36.0A	18.0B		7.28A	4.78B		8.21A	7.20A	

§The values followed by different lowercase (interaction of F × S) and upper case (Main effect of F or S) letters in a column/row are significant according to Duncan's Multiple Range Test at P=0.05, F- Fertilization, S- Soil , FYM- Farmyard manure, JNGD- Junagadh, PRBN- Parbhani

3.3. Maximum carbon carrying capacity (C_m) and C saturation deficit (S_d)

3.3.1 Alfisols

The rate of change in SOC over time in years (dC_s/dt) in Alfisol-PTMB was highest in 100%NPK + FYM (0.75 Mg ha^{-1} yr^{-1}) followed by 100%NPK+Lime (0.42 Mg ha^{-1} yr^{-1}), 150%NPK (0.32 Mg ha^{-1} yr^{-1}), 100%NPK (0.24 Mg ha^{-1} yr^{-1}), 50%NPK (0.13 Mg ha^{-1} yr^{-1}) and control (0.004 Mg ha^{-1} yr^{-1}) (supplementary Table S6). The dC_s/dt in Alfisol-BHNS were 0.66, 0.28, 0.26, 0.21, 0.12 and 0.01 Mg ha^{-1} yr^{-1}in 100%NPK+FYM, 100%NPK+Lime, 150%NPK, 100%NPK, 50%NPK and control, respectively. The dC_s/dt was invariably higher in Alfisol-PTMB than Alfisol-BHNS (supplementary Table S6). The dC_s/dt (Mg ha^{-1} yr^{-1}, y-axis) was plotted with annual average

MAERC input (Mg ha^{-1} yr^{-1}, x-axis) at 0-30 cm depth for estimating decay rate constant (*k*) (Fig.1a & b). Equating the regression equation as developed for Alfisol-PTMB (y= 0.17 x - 0.25, R^2=0.94) with Jenkinson's equation for a two C pool model, $dC_s/dt = hA - kC_s$, it is possible to estimate the humification rate constant, "*h"*, as 0.17 derived from slope of the regression line, while the decay rate constant, "*k"* as $0.25/C_s$ (Intercept/C_s), where C_s was SOC content at the beginning of the experiment (Table 5). The *h* value indicates that 17% of MAERC was annually humified and entered the SOC pool. The intercept, 0.25 Mgha^{-1} yr^{-1} of the equation represents annual loss of C from native SOM. If we equate the above value of intercept with k × Cs of Jenkinson's equation (1988) and setting the initial SOC content of 24.75 Mg ha^{-1}, the decay rate constant (*k*) of native SOC turned out as 0.01 (Table 5). This means, the C loss from native SOC over last 18 years of cultivation was 1% of the initial SOC content. By equating dC_s/dt to 0, it was possible to estimate annual C input required to maintain SOC at equilibrium (A_E) as 1.84 Mg ha^{-1} yr^{-1}. By substituting the value of *k*, C_t and dC_t/dt, the values in the equation $dC_t/dt = I (1- Ct/C_m) - kC_t$, the C_m (the maximum C carrying capacity) was computed as 37.2 Mg ha^{-1} at 0-30 cm soil depth.

The estimated C S_d in the Alfisol-PTMB at 0-30 cm depth was highest in control (0.31). The S_d decreased from 50%NPK (0.27) to 150% NPK (0.23) and 100%NPK + Lime (0.18) and 100%NPK+FYM showed the lowest value of S_d (0.05) (Fig. 2).

The relationship between dC_s/dt versus MAERC in Alfisol-BHNS is represented by the regression equation (y=0.13x – 0.04, R^2 = 0.93). Following the above analogy, the "*h*" and "*k*" in Alfisol-BHNS were 0.13 yr^{-1} and 0.003 yr^{-1}, respectively. The "*h*" represents 13% of MAERC annually humified and entered the SOC pool. The k indicates that 0.3% of C was lost from initial native SOC over 12 years of cultivation. The intercept, 0.04 Mg ha^{-1} yr^{-1}signifies annual loss of C from

native soil organic matter. The A_E was estimated as 0.39 Mg ha^{-1} yr^{-1}. The C_m in Alfisol-BHNS was computed as 21.7 Mg ha^{-1} to a soil depth of 0-30 cm.

The S_d in Alfisol-BHNS was highest in control (0.37) which decreased rapidly when the rate of fertilization increased from 50% NPK (0.31) to 100% NPK (0.33), 150%NPK (0.29), 100%NPK+FYM (0.05) and 100% NPK + Lime (0.26) (Fig. 2).

3.3.2 Vertisols

The rate of change in SOC over time in years (dC_s/dt) in Vertisol-JNGD was highest in 100%NPK + FYM (1.15 Mg ha^{-1} yr^{-1}) followed by 150%NPK (0.72 Mg ha^{-1} yr^{-1}), 100%NPK (0.40 Mg $ha^{-1}yr^{-1}$), 50%NPK (0.16 Mg ha^{-1} yr^{-1}) and control (0.02 Mg ha^{-1} yr^{-1}) (supplementary Table S6). The dC_s/dt in Vertisol-PRBN was 0.56, 0.23, 0.15, 0.12 and 0.06 Mgha^{-1} yr^{-1}in 100%NPK+FYM, 150%NPK, 100%NPK, 50%NPK and control, respectively. The dC_s/dt was higher in Vertisol-JNGD than Vertisol-PRBN (supplementary Table S6).

The relationship between dC_s/dt with MAERC in both the Vertisol-JNGD ($y=0.23x -0.30$, $R^2=0.80$) and Vertisol-PRBN ($y=0.11x - 0.12$, $R^2=0.98$) were developed (Fig. 1c & d). Like Alfisols, the "*h*", "*k*", A_E and C_m were also estimated for the Vertisols. In Vertisol-JNGD, the values of h, k, A_E and C_m were 0.23 yr^{-1}, 0.01 yr^{-1}, 1.57 Mg ha^{-1} yr^{-1} and47.6 Mg ha^{-1}, respectively (Table 5). The "h indicates that 23% of the MAERC was humified and entered the slowly decaying pool of C. The "*k*" value indicates that 10% loss of initial level of SOC over 18 years of cultivation. The amount of C input required to maintain SOC at equilibrium was estimated to be 1.57 Mg ha^{-1}. The S_d in Vertisol-JNGD decreased rapidly from control (0.36) to 100%NPK+FYM (0.08) (Fig. 2). In Vertisol-PRBN, the values of h, k, A_E and C_m were 0.11 yr^{-1}, 0.01 yr^{-1}, 0.90 Mg ha^{-1} $yr^{-1}$and

488 25.0 Mg ha^{-1}, respectively (Table 5). The 11% of MAERC was humified and entered the SOC
489 pool. The C loss from native SOC over 18 years of cultivation was 1% (*k*=0.01). The annual C
490 input required to maintain SOC at equilibrium is estimated to be 0.90 Mg ha^{-1} yr^{-1}. The S_d in
491 Vertisol-PRBN decreased from 0.40 in control to 0.36, 0.33, 0.27 in 50% NPK, 100% NPK and
492 150% NPK respectively and the S_d in 100% NPK + FYM was lowest (0.04) (Fig. 2).

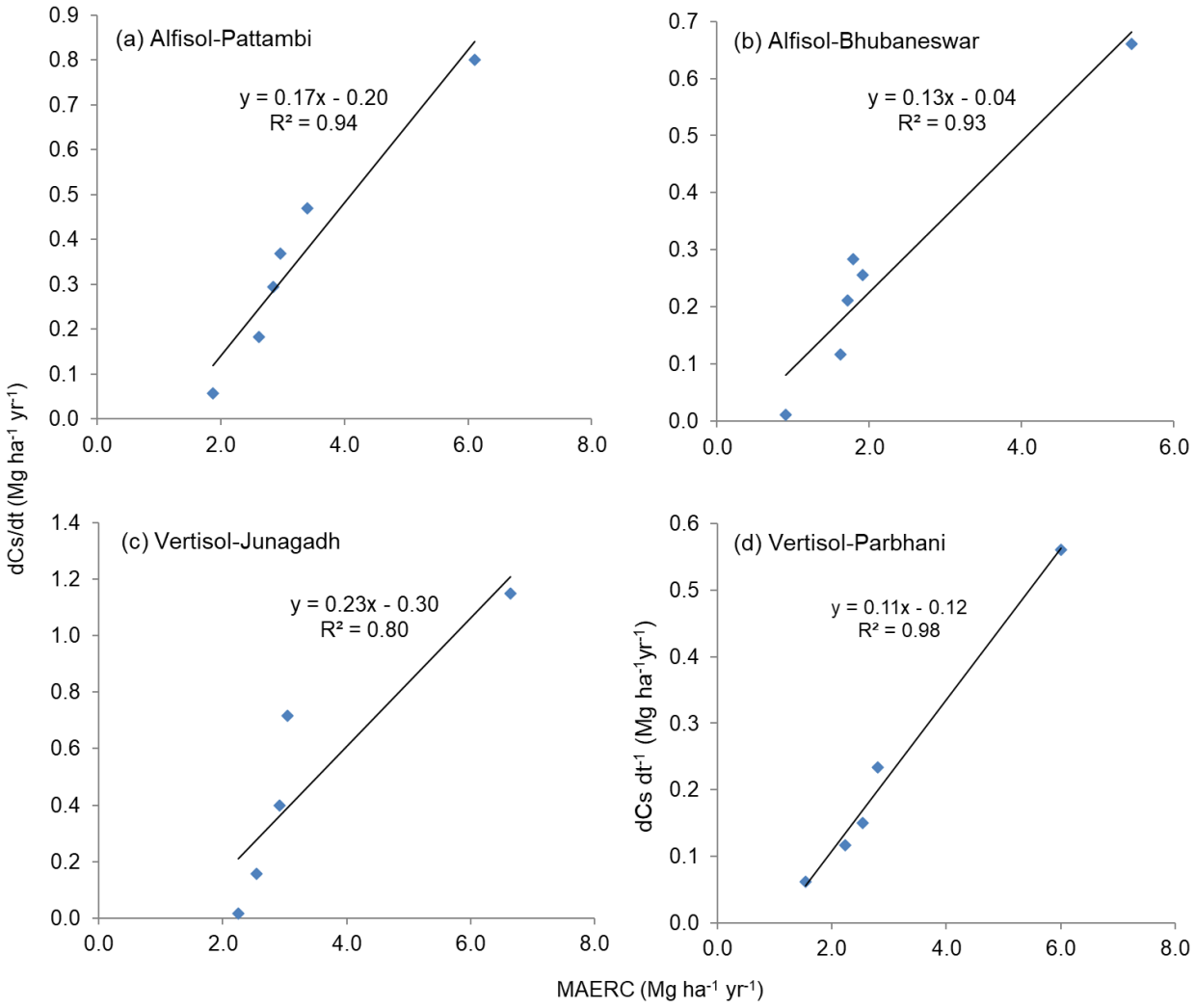
493 **Table 5**

494 Humification rate constant (h), decay rate constant (k), annual C inputs to maintain SOC at
495 equilibrium (A_E) and maximum C carrying capacity (C_m) of four soils

Soil	h (yr^{-1})	k (yr^{-1})	A_E (Mg ha^{-1} yr^{-1})	C_m (Mg ha^{-1})
Alfisol-PTMB	0.17	0.01	1.84	39.3
Alfisol-BHNS	0.13	0.003	0.39	21.7
Vertisol-JNGD	0.23	0.01	1.57	47.6
Vertisol-PRBN	0.11	0.009	0.90	25.0

496 PTMB-Pattambi, BHNS-Bhubaneswar, JNGD-Junagadh, PRBN-Parbhani

497



498

499 **Fig. 1. Relationship between mean annual estimated return of C (MAERC) with mean**
500 **annual change in SOC (dCs dt^{-1}) in Alfisols (a & b) and Vertisols (c & d)**

501

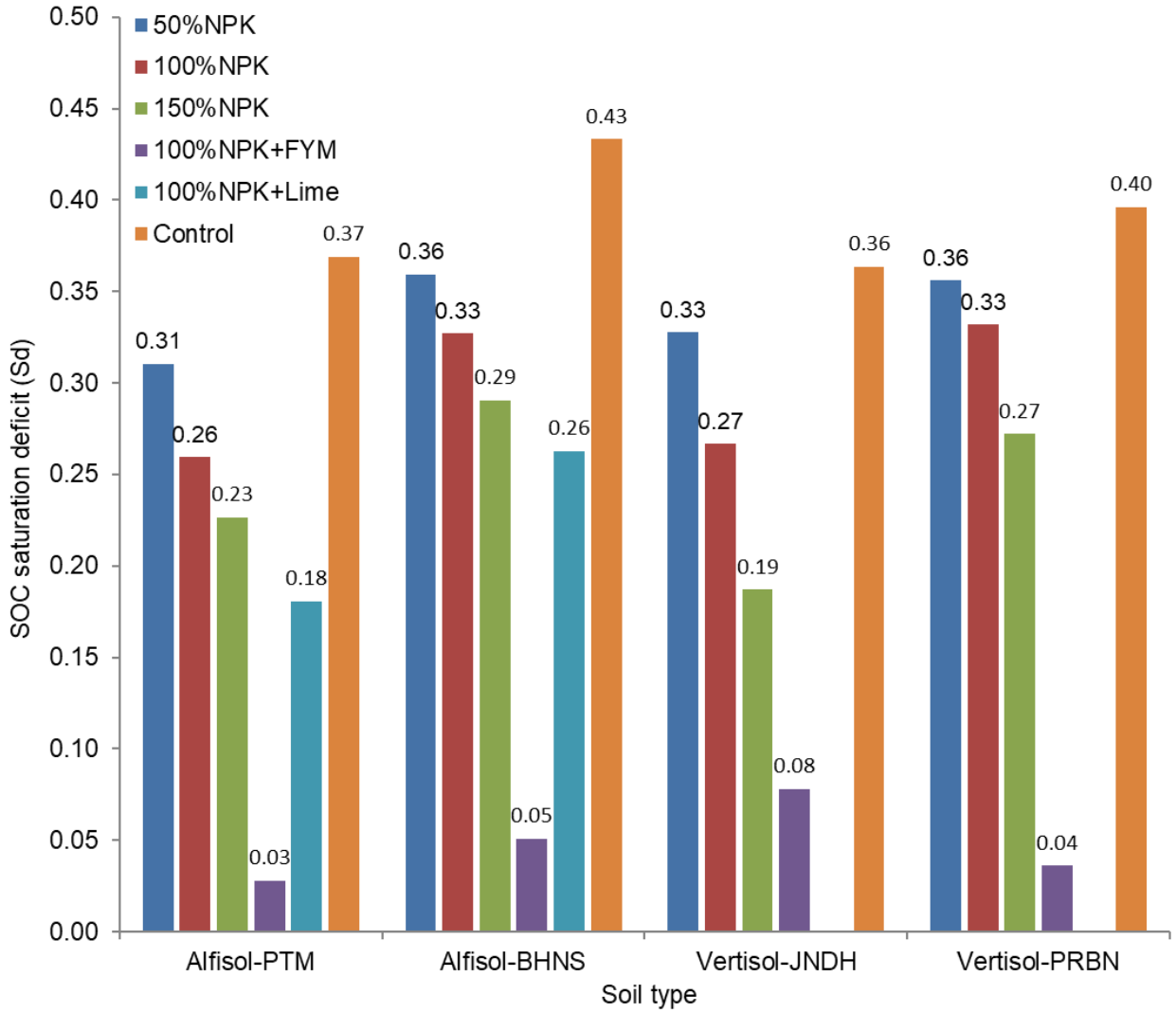


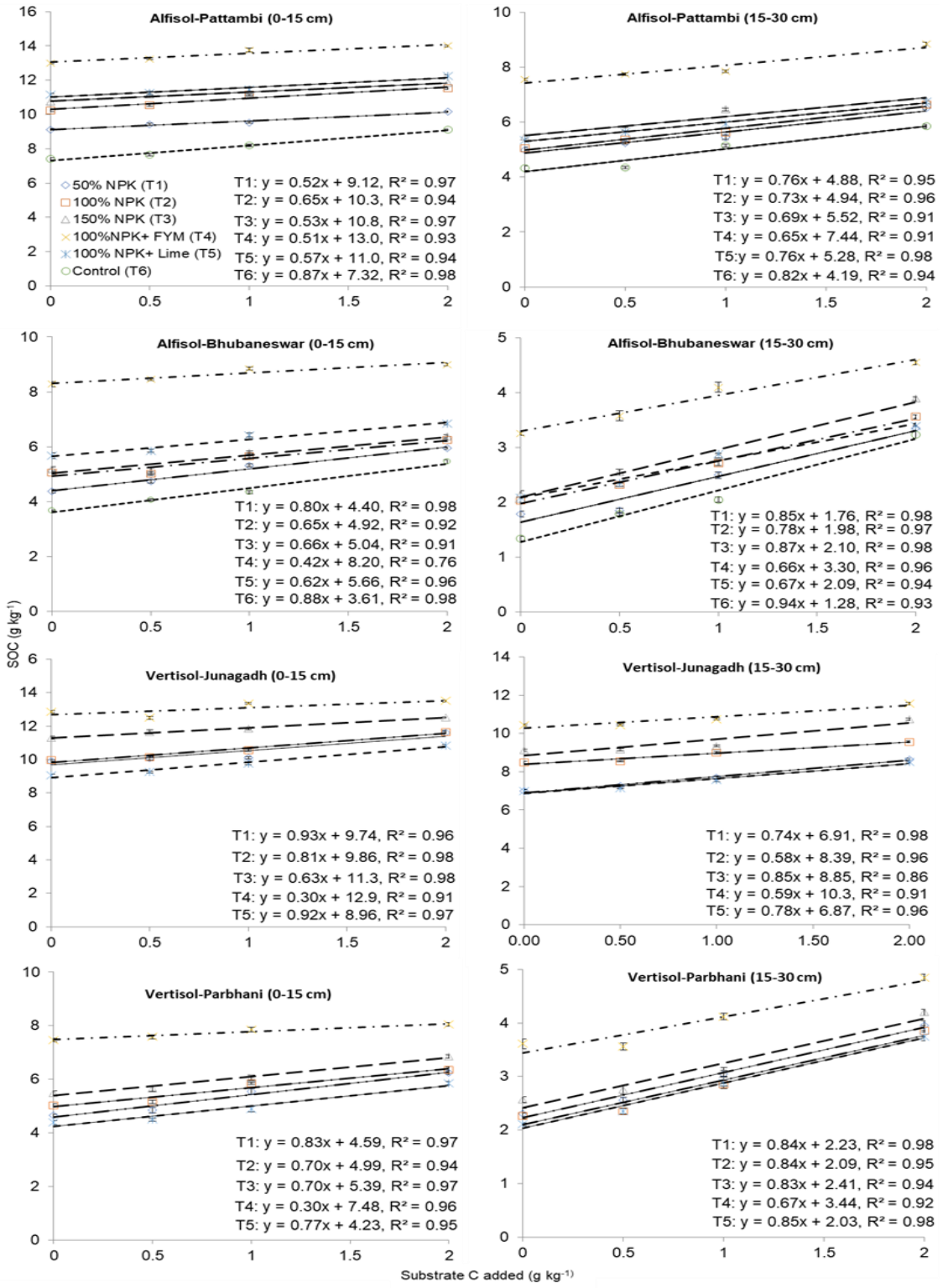
Fig. 2. SOC saturation deficits (Sd) of different treatments in Alfisols and Vertisols

3.4. Relationship between rate of change of SOC per unit substrate C and C saturation deficit (S_d)

The data pertaining to change in SOC per unit addition of substrate C (slope, dy/dx) in all the soil type at 0-15 and 15-30 cm soil depths were successfully fitted to straight line relationships (Fig. 3). Irrespective of soil orders and depths, the slope was invariably highest in the control treatment. The slope decreased rapidly when the rate of fertilization increased from 50% NPK to 150% NPK. It is interesting to note that 100% NPK+FYM showed the lowest slope among all the treatments.

In Alfisols, the 100% NPK+Lime treatment showed similar slope as that of the fertilization treatment without FYM. The average slope varied between 0.61 in Alfisol-PTMB and 0.72 in Vertisol-JNGD and in 15-30 cm soil depth the average slope value ranged from 0.71 in Vertisol-JNGD to 0.81 in the Vertisol-PRBN. On the average the slope of the straight line was higher in 15-30 cm than in 0-15 cm soil depth in all soil orders except the Vertisol-JNGD which showed almost similar slope in both soil depths. In most of the cases, the R^2 values were more than 0.90 which implies that more than 90 per cent of cases could be explained by the numerical straight-line relationship.

The correlation study revealed that the S_d computed from the carbon saturation model correlated positively with the slope of the straight line (dy/dx) developed from the substrate C driven saturation experiment. The correlation coefficient (r) was significant in the case of the Vertisol-PRBN (r=0.97), Alfisol-BHNS (r=0.93), and Vertisol-JNGD (r=0.93) (supplementary Table S7).



522

523 **Fig. 3. Relationship between changes of SOC with varying levels of substrate C added in Alfisols**

524 **and Vertisols. Error bars (±) show standard error of mean.**

525

526 **4. Discussion**

527 *4.1. Mean annual estimated return of carbon (MAERC) in soil*

528 The yield of crops is directly linked to balanced and imbalanced use of fertilizers, and optimal,
529 sub-optimal and super optimal doses of major nutrient especially N, P and K (Purakayastha et al.,
530 2008; Srinivasarao et al., 2012a). Irrespective of the locations, the MAERC was recorded highest
531 in 100%NPK+FYM. The MAERC across all t soil types followed the decreasing order:
532 100%NPK+FYM>150%NPK/100%NPK+Lime>100%NPK> 50%NPK>control. The MAERC is
533 directly related to yield of the crops which is directly linked with production of root biomass
534 triggering exudation of carbonaceous materials in the rhizosphere. The yield of crops is directly
535 linked to balanced and imbalanced use of fertilizers, optimal, sub-optimal and super optimal doses
536 of major nutrient especially N, P and K (Purakayastha et al., 2008; Srinivasarao et al., 2012a).
537 Therefore, balanced application of N, P, and K or integrated application of N, P, and K with FYM
538 showed higher yield which resulted in increased below ground biomass and rhizodeposition.
539 Purakayastha et al. (2008) also reported a similar trend in an Inceptisol under a maize-wheat-
540 cowpea cropping system under semi-arid subtropical India. The application of lime had a
541 reclamation effect in enhancing pH of acid soil to neutrality and created a congenial soil chemical,
542 physical and biological environment required for plant growth. There is a strong association
543 between the variations in crop residue C inputs into soils, which is one of the major sources of C
544 into the soil, and yield (Datta et al., 2018). Among the rice-rice cropping systems in two Alfisols,
545 the MAERC in Alfisol-PTMB was 1.65 times higher than in Alfisol from Bhubaneswar (Alfisol-
546 BHNS) and this could probably be due to higher yield realized in the former than the latter soil.
547 Our study corroborated the findings of Mandal et al. (2008) who reported 9.55 Mg ha^{-1} yr^{-1} C input

derived from the NPK + compost treatment in rice-based cropping systems and they linked this phenomenon with higher rice yield in this treatment. The Vertisol-JNGD showed 1.15 times higher MAERC than Vertisol-PRBN. The MAERC in 100% NPK + FYM in Vertisol-PRBN under soybean-safflower system was 6.01 Mg ha^{-1} yr^{-1} which was higher than the reported value of 4.97 Mg ha^{-1} yr^{-1} in a similar type of soil under sorghum-wheat cropping system (Datta et al., 2018). The higher MAERC in the soybean-safflower system than the sorghum-wheat system could be due to higher return of residues, roots, and nodules to the soil. The addition of matured and stable FYM with low C:N with carbonaceous material having high C:N had a priming effect on faster decomposition augmenting C cycling processes which caused release of nutrients essential for plant growth and faster humification in soil (Purakayastha et al., 2019). Overall, the Vertisols had higher MAERC than Alfisols. This could be attributed to higher biomass yield in the former soil than the latter, effect of crop residues (leguminous crop in the latter) and difference in clay minerology (dominated by 2:1 in the latter) at the studied sites (Singh et al., 2018).

4.2. Sequestration and stabilization of soil organic carbon

The higher SOC stock in Alfisol-PTMB (31.3 Mg ha^{-1}) than Alfisol-BHNS (16.7 Mg ha^{-1}) was mainly due to higher MAERC in the former than the latter soil. Besides this, Alfisol-PTMB being sandy clay loam in texture retained more of C than the Alfisol-BHNS, being sandy loam in texture, though both the soils have almost similar 1:1 type kaolinite clay mineralogical make up. When the treatments were compared, it was observed that the integrated use of NPK with FYM showed higher amount of C input over sub-optimal, optimal, or super optimal doses of NPK. The application of organic matter such as FYM, rice straw and green manure increased the build-up of SOC stock in rice-based cropping systems at tropical and subtropical Asian countries

(Bhattacharyya and Pal., 2003). Our results corroborated the findings of Srinivasarao et al. (2012b) who reported that the application of FYM alone or in combination with chemical fertilizers significantly contributed to higher amounts of C inputs which enriched SOC content by 41 to 73% over 13 years in groundnut- finger-millet crop rotation in Alfisol of Bengaluru, India.

The stabilization of SOC in Alfisol-PTMB was significantly higher than in Alfisol-BHNS. The slow and incomplete decomposition of OM under anaerobic condition of the rice based cropping system in two Alfisols of our study might have contributed significantly to higher accumulation and stabilization of C (Bhattacharyya et al., 2007). Srinivasarao et al. (2012b) observed that 36.8 % of C applied as FYM was stabilized compared to the control and mere NPK treatments.

The groundnut-wheat cropping system in Vertisol-JNGD and soybean–safflower system from Vertisol-PRBN recorded highest SOC stock in 100% NPK+ FYM treatment. The SOC stocks in these soils (43.9 Mg ha^{-1} in Junagadh, and 24.1 Mg ha^{-1} in Parbhani) were well associated with the MAERC values in these sites, i.e., 6.65 Mg $ha^{-1}yr^{-1}$ in Junagadh and 6.01 Mg ha^{-1} yr^{-1} in Parbhani. Srinivasarao et al. (2012a) reported that the combined application of FYM with chemical fertilizers increased SOC stock to 69.9 Mg $ha^{-1}$and increased the C sequestered to 11.9 Mg ha^{-1} yr^{-1} compared to control and unfertilized treatments in 15 years of a long-term experiment conducted in soybean-safflower in a Vertisol. The long-term application of fertilizers in combination with manures was found to increase SOC stock by 12-113% by many researchers in other parts of the world (Chai et al., 2015; Zhang et al., 2015). The positive effect of application of manure on yield and thereby SOC could be attributed to improved soil health and enhanced availability of nutrients.

On an average the amount of SOC sequestered was significantly higher in Vertisol-JNGD (7.28 Mg ha^{-1}) than in Vertisol-PRBN (4.78 Mg ha^{-1}). In this connection, Lakaria et al. (2012) reported

a significant increase in SOC built-up under soybean–wheat cropping system in Vertisol with long term fertilization. Guo et al. (2019) observed that continuous fertilization for 34 years, increased SOC sequestration by 0.66 Mg ha^{-1} yr^{-1} in the combined fertilizer + manure treatment compared to 0.08 Mg ha^{-1} yr^{-1} in the control treatment. Similar to our findings, Gou et al. (2019) also reported that the continuous applications of organic fertilizers increased the amount of C sequestered in the Vertisol by 16-132% compared to inorganic fertilization. The increase in C storage from organic fertilizer could be attributed to the direct C input from the manure and thereby enhanced biomass production (Maillard and Angers, 2014; Das et al., 2019).The stability of soil C is also affected by the different biochemical constituents of organic matter owing to change in the microbial community (Mohanty et al., 2013; Baumann et al., 2009; Das et al., 2019).

4.3. Humification rate constant (h) and decay rate constant (k)

The humification rate constant (h) was marginally higher in Alfisol-PTMB (0.17) than Alfisol-BHNS (0.13). This could be one of the reasons behind higher conversion of input C into stable C in Alfisol-PTMB than Alfisol-BHNS by humification process. The h was close to 0.20 yr^{-1} as reported from other cultivated systems (Paustian et al., 1997; Huggins et al., 1998b; Purakayastha et al., 2008). The annual loss of C from native soil organic matter was observed to be 0.20 Mg ha^{-1} yr^{-1} in Alfisol-PTMB, and 0.04 Mg ha^{-1} yr^{-1} in Alfisol-BHNS. This can be explained by the decay rate constant (k) value which was higher in Alfisol-PTMB (0.01) than in Alfisol-BHNS (0.003). In a long-term fertilizer experiment, Kundu et al. (2007) also reported a low k value (0.0018 yr^{-1}) in a Typic Haplaquept of Indian Himalayas.

Among the Vertisols, the humification rate constant Vertisol-JNGD was higher than Vertisol-PRBN with an h value of 0.23 yr^{-1} in the former and 0.11 yr^{-1} in the latter. The decay rate constant (k) in Vertisol-JNGD and in Vertisol-PRBN was 0.01. The stabilization of SOC in these Vertisols occurs due to 2:1 type of expanding lattice clay minerals. Soils rich in 2:1 clay minerals, e.g., montmorillonites and vermiculites, have a greater ability to protect the C in soil than 1:1 clay minerals, e.g., kaolinites (Six et al., 2002), due to their higher specific surface area and CEC (Hassink, 1997; Wattel-Koekkoek and Baumann ., 2004). The two Vertisols having the same clay mineralogy thus showed similar SOC stabilization. When all the four soils were compared, it was observed that not only the clay mineralogy but also other factors like growing conditions (aerobic versus anaerobic), climate, and litter quality also played a significant role in stabilization of SOC. Litter quality with respect to lignin content and C:N influences decomposability, and generally residues with relatively high lignin content and higher lignin +polyphenol ratio and lower C:N ratio decompose rapidly (Meentemeyer, 1978; Laishram and Yadava, 1988). Legumes are richer in protein with narrow C:N, which drives microbial activity. During decomposition, residue C chemistry, mainly the relative content of aryl-C and alkyl-C and microbial community structure changed over time and differed between residue types (Baumann et al., 2009). Thus, in the legume-oilseed based cropping system (soybean-safflower) in Vertisol-PRBN, the left-over litter was easily degradable as evidenced by higher *k* which showed lowest SOC stabilization. The Vertisol-PRBN though dominated by smectic clay minerals but the humification rate constant was lowest and on the other hand Alfisols dominated by kaolinitic clay minerals showed higher "*h"* than the Vertisol-PRBN. The aerobic-anaerobic cycle in rice-rice cropping system in both the Alfisols regulated the decomposition of added C leading to higher "*h"* and lower *k* than Vertisol-PRBN. The aerobic-anaerobic cycle in rice-rice system was reported to limit the oxidation of SOC

resulting in more accumulation of OC (Yadav et al., 2017). On the contrary, clay mineralogy played a dominant role in Vertisol-JNGD especially under moderate (oilseed) to inferior (cereal) litter quality which favored highest "*h"* and lowest "*k"*, thus showed highest SOC stabilization. This observation is supported by the fact that combined application of green manure, FYM and biofertilizer was reported to exhibit highest humification and aromaticity in Inceptisol of semi-arid subtropical India (Purakayastha et al., 2019).

4.4. Maximum carbon carrying capacity (C_m), carbon saturation deficit (S_d) and carbon input to maintain soil organic carbon at equilibrium (A_E)

The maximum C carrying capacity (C_m) in Alfisol from Pattambi (Alfisol-PTMB) (39.3 Mg ha^{-1}) was almost twice that of Alfisol from Bhubaneswar (Alfisol-BHNS) (21.7 Mg ha^{-1}). Among the Vertisols, the C_m in Vertisol-JNGD was estimated to be 47.6 Mg ha^{-1}, which is almost two times higher than that of Vertisol-PRBN (25Mg ha^{-1}). Our results corroborated the findings of Kundu et al. (2001) who also reported that annual rate of change in SOC stock at 0-30 cm soil depth correlated positively with gross annual C input. The C_m in the Alfisols was lower comparatively, mainly because of its distribution in the tropical subhumid and perhumid regions, where the mean annual rainfall was between 1449 and 2727 mm. Some of the important factors affecting the sequestration of C in soil are temperature and rainfall as they play an important role in biomass production (Singh et al., 2018). The availability of adequate moisture content may not accelerate the degradation of OM with increase in temperature (Singh et al., 2017). The relatively lower content of SOC in the warmer regions is mainly caused by increased microbial activity (Nishina et al., 2014; Lal, 2007). Moreover, the amount and content of clay also influence the C_m. The clay

content in the Alfisols (22 to 28%) is low compared to that in the Vertisol (76%) and is mainly dominated by 1:1 type of clay minerals. The average S_d in Vertisol-JNGD and Vertisol-PRBN were 0.24 and 0.28, respectively. Overall, the C_m was higher in Vertisols than Alfisols. The clay mineralogical make up and swelling and shrinking properties of Vertisols strongly influences the SOM dynamics significantly (Hidialgo et al., 2019; Somasundaram et al., 2017; Lopez-Bellido et al., 2016). The positive effect of 2:1 expanding clay mineral on the sequestration and stabilization of soil C under tropical climate in India was highlighted by Das et al.(2023). The higher clay content in the Vertisols facilitated enhanced protection of soil organic matter (SOM) and thereby to greater structural stabilization. Besides the clay content, type of clay, climate of the region, crop growing conditions and quality of residues also play a significant role in SOM dynamics. It is supported by the fact that the temperature sensitivity of SOM mineralization is directly influenced by clay mineralogy (Sarkar et al., 2018). With rise in temperature, SOM held by weak forces would be released into the soil solution in labile forms, which gets subjected to mineralization, whereas, those held by strong forces, get released slowly into the soil solution and thereby are protected against rapid mineralization. Changes in moisture contents are likely to have a positive or a negative effect on the C sequestration capacity of soils (Singh et al., 2017). In wetter soils, higher microbial decomposition is observed in the presence of good air movement, in contrast to drier soils (Moyano et al., 2013). However, the SOC content in the humid region is higher than that of the drier region, which can be attributed to the higher net primary productivity in the humid region compared to drier regions (Lal, 2007).

The C saturation deficit (S_d) was invariably higher in the control treatment than in the other treatments across all soil types. The SOC content in this treatment was far away from the C_m and, therefore, this treatment showed the highest S_d. Interestingly, irrespective of soil type, S_d by and

large varied between 0.03 to 0.43. This indicates that without any fertilization or manuring the soil can maintain 3 to 43% of C_m which implies that C input especially from left over shoot and root biomass, and rhizodeposition as shown in the MAERC data might have contributed to the SOC pool. In contrast to the control treatment, other treatments showed variable S_d values between soil types, e.g., Alfisol-PTMB/Vertisol-JNGD showed lower S_d than Alfisol-BHNS/Vertisol-PRBN. This phenomenon implies that long-term fertilization helped Alfisol-PTMB to approach more towards C_m than Alfisol-BHNS. Similar analogy is also applicable for Vertisol-JNGD which was closer to saturation than Vertisol-PRBN. The higher SOC sequestered (6.55 Mg ha^{-1}) in the Alfisol- PTMB compared to that of Alfisol-BHNS (4.62 Mg ha^{-1}), reflects the lower S_d in the former soil. Similar observations were also recorded in Vertisols, where the higher SOC sequestered (7.28 Mg ha^{-1}) in Vertisol-JNGD, reflects a comparatively lower S_d than in Vertisol-PRBN (4.78 Mg ha^{-1}). The study clearly demonstrated that proper management strategies e.g., liming of acid soil, balanced fertilization with NPK, optimal/super optimal doses of NPK and integrated use of fertilizers and manures (100%NPK+FYM) are capable of reducing the S_d until the SOC reaches at the level of C_m. Therefore, S_d followed the decreased order: control > 50%NPK > 100%NPK > 150%NPK/100%NPK+Lime> 100%NPK+FYM. The negative S_d value in the 100% NPK+FYM treatment at both Alfisol-PTMB and Alfisol-BHNS and at Vertisol-PRBN was due to under estimation of C_m in 100%NPK+FYM in the above soils except Vertisol-JNGD by the C saturation model of Stewart et al. (2007) which was used for this study but in other treatments the S_d was positive indicating the usefulness of this model. This suggests that the C saturation model of Stewart et al. (2007) needs further improvement while estimating C_m in the soils under study.

704 The slope (dy/dx, change in SOC per unit change in added substrate C) calculated from the
705 incubation experiment was highest in the control treatment in all the soil orders (Fig.3). We
706 hypothesized that the soil away from the C saturation or C carrying capacity possesses higher CSP.
707 Our study clearly established that 100%NPK+FYM treatment showed lowest S_d and therefore it
708 exhibited lower rate of SOC increment per unit substrate C added. Other sub-optimal (50%NPK),
709 optimal (100%NPK) and super-optimal (150%NPK) doses of NPK showed gradual decrease in S_d
710 (Fig. 2) and thereby decreased the slope of the straight line (Fig. 3). The slope of the line in general
711 was higher in lower depth than the surface depth which signifies higher C enrichment potential of
712 the former than the latter. As the slope of the straight line in Alfisol-BHNS and Vertisol-PRBN
713 was higher than Alfisol-PTMB and Vertisol-JNGD, the former soil showed higher CSP than the
714 latter soil. This is explained by the fact that Alfisol-BHNS and Vertisol-PRBN showed relatively
715 higher S_d than the Alfisol-PTMB and Vertisol-JNGD (Fig. 3). Stewart et al. (2008) also conducted
716 incubation studies with ^{13}C labelled residues for 2.5 years and observed that the amount of C
717 stabilized was related to S_d and those soils with low C content and under degradation had greater
718 potential to sequester more C because of higher S_d. The higher slope in the control and 50% NPK
719 treatments indicated that the addition of different doses of substrate C were not able to saturate
720 these soils which resulted in a linear increase of the SOC contents linearly. The results derived
721 from the incubation study aligned with that obtained from the C saturation model. The positive
722 correlation between S_d computed by the C saturation model and the slope calculated from the
723 incubation study well associated with each other.

724 The annual C inputs required to maintain SOC at equilibrium (A_E) in Alfisol-PTMB (1.84 Mg ha^{-1}
725 yr^{-1}) was four and a-half times higher than that required for Alfisol-BHNS (0.39 Mg ha^{-1} yr^{-1}).
726 As the initial level of SOC stock in Alfisol-PTMB was almost double that of Alfisol-BHNS, more

C input is required to maintain the sustenance level of C in native SOC pool. The A_E in Vertisol-PRBN was 0.90 Mg ha^{-1} yr^{-1}, compared to 1.57 Mg ha^{-1} yr^{-1}in Vertisol-JNGD. In another study, Datta et al. (2018) reported that a critical amount of 0.96 Mg ha^{-1} yr^{-1} C input needed to be incorporated in the soil to maintain the existing SOC level, zero change, for the long-term sustenance of sorghum-wheat production system in the semi-arid Vertisols. A critical amount of 1.1 Mg C $ha^{-1}yr^{-1}$ was reported by Srinivasarao et al. (2012a) in Vertisols for sorghum mono-crop system. Similar observations were reported by Majumder et al. (2007) in alluvial soil under rice-wheat-jute system (4.6 Mg ha^{-1} yr^{-1}) and Mandal et al. (2007) for a rice-based system (2.9 Mg $ha^{-1}yr^{-1}$) in subtropical regions of India. The above observation suggests that A_E varies considerably depending upon the soil types, cropping systems, climatic conditions etc.

5. Conclusions

The hypothesis set for the study was confirmed when all the factors were taken into consideration. Though clay mineralogy and texture are important factors for SOC stabilization but may not always be true and therefore other factors like, growing conditions (aerobic versus anaerobic), litter chemistry, climate are also equally important which should be considered while formulating strategies for soil C sequestration. The soil organic carbon (SOC) sequestration, maximum C carrying capacity (C_m), and C S_d in Alfisol and Vertisol varied significantly due to several factors. Of them, soil texture, mean annual estimated return of C (MAERC) and climate largely influenced the C sequestration and C_m in Alfisol.In Vertisol, MAERC and climate played the most crucial role. Increase in soil aridity as in the case of Vertisol-PRBN decreased the CSP. Vertisol-PRBN in spite of being clayey in texture and dominated by 2:1 type smectite clays showed lower SOC stock and C_m than Alfisol-PTMB that was sandy loam in texture and dominated by 1:1 type

kaolinite clay mineral. Thus, soil texture, clay mineralogy, C input, climate and crop growing conditions interacted in a complex way to stabilize C in the studied soils. The integrated application of 100%NPK with either lime (for acidic Alfisols) or FYM is recommended for higher stock, sequestration and stabilization of SOC. The C saturation deficit (S_d) and rate of change in SOC per unit addition of substrate C significantly correlated with each other implying a greater C sequestration in those soils which had a lower SOC level. The C S_d narrowed down in the treatment 100%NPK+FYM due to its progress towards C saturation level of soil. Therefore, blanket recommendation of FYM needs to be modified depending upon the C saturation level of soil and assessing the minimum C input required to maintain SOC at equilibrium. Otherwise, the conventional practice of blanket recommendation might release a large amount of C in the form of CO_2, a greenhouse gas responsible for global warming. The higher humification rate constant (h) in Alfisol-PTMB than Alfisol-BHNS indicated a higher conversion of input C into stable C in Alfisol-PTMB than Alfisol-BHNS by the humification process. The minimum threshold limit of annual C inputs (A_E) at the rate of 1.84 and 0.39 Mg ha^{-1} yr^{-1} are recommended for Alfisol-PTMB and Alfisol-BHNS, respectively, to protect and preserve the native SOC. The corresponding A_E values for Vertisol-JNGD and Vertisol-PRBN are 1.57 and 0.90 Mg ha^{-1} yr^{-1}. Shifting to high fertility treatments (100%NPK+FYM), the CSP in Alfisol and Vertisol occupying an area 26 and 42 Mha in India was estimated to be 31 and 22 Tg C yr^{-1}, which is equivalent to 5 and 3.5% of annual greenhouse gas emissions [CO_2-C equivalent (2307 Mt CO_2e], respectively.

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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Authors' contributions

SS and TJP proposed the concept, hypothesis and objectives of the work in on-going long-term fertilizer experiments at multiple locations that are managed by MS, TV, KKR, STS, MS. SS analyzed all the samples, did statistical analysis of the data, and written the first draft of the paper in consultation with TJP and BS. TJP, BC, BS, KKB edited the subsequent drafts and significantly contributed to result and discussion. MS, TV, KKR, STS and MS recorded all yield data of crops and provided logistical support for collection of soil samples.

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1014

Supplementary Information

Table S1 Details of experimental sites located in different states of India

Location	Pattambi (Kerala state)	Bhubaneswar (Odisha state)	Junagadh (Gujarat state)	Parbhani (Maharashtra state)
Climate	Per-humid	Tropical and sub-humid	Dry sub-humid	Semi-arid
Agro ecological sub-region	Central and South Sahyadris, hot moist sub-humid to humid transitional	Eastern Ghats, hot moist sub-humid	South Kachchh and North Kathiawar Peninsula, hot moist arid	Central and Western Maharashtra Plateau and North Karnataka Plateau and North Western Telangana Plateau, hot moist semi-arid
Year of commencement	1996	2002	1996	1996
Altitude (m, msl)	32	30	107	347
Latitude	10 º 48’ 40.5” N	20º 15’ 54.1” N	21º29’ 59.7” N	19 º14’ 34.7” N
Longitude	76 º 11’ 23.3” E	85 º 48’ 42.2” E	70º26’ 52.6” E	76 º47’19.2” E
Temperature (ºC)	32	33	28.3	26.1
Mean annual rainfall (mm)	2727	1449	844	874
Cropping systems	Rice-rice	Rice-rice	Ground-wheat	Soybean-safflower
Fertilizer dose (N-P-K, kg ha^{-1})	90-20-38 (both the rice)	100-26-50 (Both the rice)	25-22-0 (Groundnut) 120-26-50 (Wheat)	30-26-25 (Soybean) 60-18-0 (Safflower)
Manure doses (Mg ha^{-1})	5 Mg ha^{-1}/ season	5 Mg ha^{-1}/ season	5 Mg ha^{-1}/ season	5 Mg ha^{-1}/ season
Puddling/plowing intensity	4 times /annum	4 times /annum	4 times /annum	4 times /annum

Table S2

Physico-chemical and chemical characteristics of the experimental soils

Parameter	Alfisol-PTMB	Alfisol-BHNS	Vertisol-JNGD	Vertisol-PRBN
Mechanical analysis				
Sand (%)	62.2	66.24	17.9	14.6
Silt (%)	10.3	12.28	6.1	9.96
Clay (%)	27.5	21.48	76.0	75.5
Texture	Sandy clay loam	Sandy loam	Clay	Clay
Taxonomic class	Typic Haplustalf	Aeric Haplustalf	Vertic Ustochrept	Typic Chromustert
Clay Minerology	Kaolinite	Kaolinite	Smectite, Kaolinite	Smectite, Kaolinite
Physico-chemical properties				
pH	5.50	6.40	8.5	7.7
EC(ds m^{-1})	0.04	0.104	0.18	0.35
Chemical properties				
SOC (g kg^{-1})	15.0	7.0	16.0	5.0
Available N (kg ha^{-1})	242	150	145	232
Available P_2O_5 (kg ha^{-1})	15.3	7.77	25.9	6.71
Available K_2O (kg ha^{-1})	80.0	163	60.0	544

PTMB-Pattambi, BHNS-Bhubaneswar, JNGD-Junagadh, PRBN-Parbhani

Table S3

Recommended Fertilizer Dose of different crops in the selected site

Location	Crop	Recommended fertilizer dose (kg ha^{-1})		
		N	P	K
Bhubaneswar	Rice (*Oryza sativa* L.) (*Kharif*)	100	26	50
	Rice (*Rabi*)	100	26	50
Pattambi	Rice (*Kharif*)	90	20	38
	Rice(Rabi)	90	20	38
Junagadh	Groundnut (*Arachis hypogeae* L.)	25	22	0
	Wheat (*Triticum aestivum* L.)	120	26	50
Parbhani	Soybean (*Glycine max* L.)	30	26	25
	Safflower (*Carthamus tinctorius* L.)	60	18	0

Table S4

Bulk density of four soils at 0-15 and 15-30 cm depths

Treatments	Alfisol				Vertisol			
	Pattambi		Bhubaneswar		Junagadh		Parbhani	
	BD (Mg m^{-3})							
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
50% NPK	1.24±0.012¶	1.28±0.014	1.34±0.009	1.36±0.015	1.19±0.009	1.20±0.01	1.34±0.009	1.40±0.009
100% NPK	1.23±0.015	1.26±0.013	1.32±0.010	1.35±0.012	1.18±0.006	1.21±0.009	1.32±0.007	1.39±0.012
150%NPK	1.21±0.009	1.24±0.011	1.33±0.007	1.36±0.012	1.18±0.009	1.21±0.006	1.31±0.015	1.37±0.006
100%NPK+FYM	1.20±0.015	1.21±0.02	1.26±0.012	1.28±0.015	1.18±0.011	1.20±0.003	1.30±0.006	1.34±0.009
100%NPK+Lime	1.26±0.013	1.27±0.006	1.32±0.012	1.32±0.009	-	-	-	-
Control	1.36±0.011	1.38±0.01	1.44±0.03	1.46±0.020	1.19±0.012	1.20±0.007	1.35±0.007	1.42±0.012

¶Mean±value indicates standard error of mean

Table S5

Mean annual estimated return of C from various sources contributing to Alfisols and Vertisols under different cropping systems

Treatments	Alfisol			Vertisol								
	Rice-Rice (Pattambi)			Rice-Rice (Bhubaneswar)			Groundnut-Wheat (Junagadh)			Soybean-Safflower (Parbhani)		
	AGBC§	BGBC	EOC	AGBC	BGBC	EOC	AGBC	BGBC	EOC	AGBC	BGBC	EOC
	(Mg ha^{-1} yr^{-1})											
50%NPK	0.371±0.02¶	2.24±0.14		0.235±0.03	1.39±0.12		0.397±0.09	2.15±0.09		0.302±0.06	1.94±0.10	
100%NPK	0.403±0.06	2.44±0.17		0.254±0.02	1.46±0.14		0.730±0.04	2.49±0.16		0.340±0.01	2.19±0.12	
150%NPK	0.428±0.03	2.53±0.14		0.275±0.05	1.64±0.23		0.450±0.12	2.59±0.07		0.373±0.03	2.43±0.14	
100%NPK+FYM	0.402±0.04	2.41±0.10	3.40±0.12	0.298±0.08	1.81±0.16	3.34±0.18	0.493±0.02	2.94±0.21	3.22±0.02	0.372±0.01	2.40±0.09	3.30±0.12
100%NPK + Lime	0.477±0.09	2.92±0.10		0.257±0.03	1.53±0.14		-	-		-	-	
Control	0.230±0.04	1.65±0.15		0.137±0.04	0.76±0.10		0.301±0.03	1.95±0.18		0.210±0.05	1.34±0.08	

§AGBC- Above Ground Biomass Carbon, BGBC- Below Ground Biomass Carbon, EOC- External Organic Amended Carbon

¶Mean±value indicates standard error of mean

Table S6

Mean annual rate of SOC change per year (dCs dt^{-1}) in four soil orders

Fertilization/Soil	dCs dt^{-1} (Mg ha^{-1} yr^{-1})			
	Alfisol-PTMB	Alfisol-BHNS	Vertisol-JNGD	Vertisol-PRBN
50% NPK	0.18	0.12	0.16	0.12
100% NPK	0.29	0.21	0.40	0.15
150% NPK	0.37	0.26	0.72	0.23
100% NPK+FYM	0.80	0.66	1.15	0.56
100%NPK+Lime	0.47	0.28	-	-
Control	0.057	0.01	0.02	0.06

PTMB-Pattambi, BHNS-Bhubaneswar, JNGD-Junagadh, PRBN-Parbhani

Table S7

Correlation coefficient (r) between saturation deficit (S_d) with rate of change in SOC content per unit addition of substrate C (dy/dx) (n=6 for Alfisol, n=5 for Vertisol)

Soil order	Parameter	Slope (dy/dx) (g SOC/g substrate C)	Saturation deficit (S_d)
Alfisol-PTMB	dy/dx	1	
	S_d	0.74^{ns}	1
Alfisol-BHNS	dy/dx	1	
	S_d	0.93**	1
Vertisol-JNGD	dy/dx	1	
	S_d	0.93*	1
Vertisol-PRBN	dy/dx	1	
	S_d	0.97**	1

* Significant at 5% level of significance

**Significant at 1% level of significance

^{ns} Not significant