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Eco-Restoration of Coal Mine Spoil: Biochar Application and Carbon Sequestration for Achieving UN Sustainable Development Goals 13 and 15

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Abstract: Open cast coal mining causes complete loss of carbon sink due to the destruction of vegetation and soil structure. In order to offset the destruction and to increase sequestration of carbon, afforestation is widely used to restore these mine spoils. The current field study was conducted to assess the ecosystem status, soil quality and C pool in an 8 years old reclaimed mine spoil (RMS), compared to a reference forest (RF) site and unamended mine spoil (UMS). Biochar (BC) prepared from invasive weed *Calotropis procera* was applied in this 8 year RMS at 30 t ha⁻¹ (BC₃₀) and 60 t ha⁻¹ (BC₆₀) to study its impact on RMS properties and C pool. Carbon fractionation was also conducted to estimate inorganic, coal and biogenic carbon pools. The C stock of 8 year old RMS was 30.98 Mg C ha⁻¹ and sequestered 113.69 Mg C ha⁻¹ CO₂. BC₃₀ and BC₆₀ improved the C-stock of RMS by 31% and 45%, respectively, and increased the recalcitrant carbon by 65% (BC₃₀) and 67% (BC₆₀). Spoil physio-chemical properties such as pH, cation exchange capacity, moisture content and bulk density were improved by biochar application. The total soil carbon at BC₃₀ (36.3 g C kg⁻¹) and BC₆₀ (40 g C kg⁻¹) was found to be significantly high compared to RMS (21 g C kg⁻¹) and comparable to RF (33 g C kg⁻¹). Thus, eco-restoration of coal mine spoil and biochar application can be effective tools for coal mine reclamation and can help in achieving the UN sustainable development goal 13 (climate action) by increasing carbon sequestration and 15 (biodiversity protection) by promoting ecosystem development.

Keywords: coal mine spoil; reclamation; biochar; carbon sequestration; carbon fractionation

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1. Introduction

The UN sustainable development goal (SDG) 13 stands for climate action and promotes all activities which would ensure successful sequestration of carbon, whereas, SDG 15 safeguards and restores biodiversity protection [1,2]. Burning of fossil fuels is the primary drivers of global warming and climate change, and the extraction of these resources also adds to global concerns regarding the climate crisis [3,4]. Mining activities lead to complete loss of vegetation and the carbon sink in the soil and plants are lost to the atmosphere [5]. Mine spoils are carbon deficit with impoverished soil conditions that cannot support plant and microbial growth. Coal mine restoration can help restore the lost carbon sink by promoting plant growth and enriching the mine spoil, which helps sequester the atmospheric carbon [6,7]. The most common techniques for mine restoration include afforestation, agriculture and grassland development [8,9]. Plantation of hardy species in reclaimed mine spoils (RMS) improves the soil organic carbon (SOC) pool and improves the carbon sequestration potential of the ecosystem [10]. Development of natural forest in mine spoils may take centuries due to the impoverished soil properties and lack of substrate for supporting plant growth. Degraded land can be reclaimed by development of forest cover. Technical reclamation such as leveling and grading of dump, reducing slope

length, stabilization of slope by blanketing with coir mat along with grass-legume mixture, application of top soil, fly ash and bio-solids can be used to enhance the vegetation growth [11–13].

Restoration of RMS can potentially enhance soil C sequestration rate and improve soil properties [14–16]. Akala and Lal, [17] reported that the SOC pool of a RMS increased from 14 Mg ha⁻¹ to 48.4 Mg ha⁻¹ after 21 years of pastureland development in a degraded coal mine site. In another 19 years old revegetated coal mine spoil (Singrauli, India) there was 712% increase in the rate of carbon sequestration [18]. The sequestration of carbon depends on the type of vegetation used for reclamation, age of reclamation and nature of coal mine spoil. A study conducted by Mukhopadhyay et al. [19] in RMS reported that the carbon density was higher for *Dalbergia sissoo* Roxb. and *Acacia auriculiformis* A. Cunn. ex Benth. (39.6–43.7 kg C tree⁻¹) and lowest for *Albizzia lebbeck* L. (20.7 kg C tree⁻¹). Thus, plantation can be an effective tool for coal mine spoil eco-restoration and enhanced carbon sequestration.

A number of studies reported an increase in the soil carbon stock by biochar application [20,21]. Biochar is a thermal degradation product of biomass produced in a pyrolysis like condition by limiting the supply of oxygen [21]. Pyrolytic conversion of biomass produces aromatic carbon that is resistant to degradation in soil, thus considered an option to address the global CO₂ emission problems by biomass decomposition [22]. Biochar has a high mean residence time and aromaticity, making it highly recalcitrant in nature [11,23]. Thus, carbon that would normally be released as CO₂ through biomass decomposition is converted to biochar which is highly stable and aromatic. The aromaticity of biochar depends on the chemistry of biomass used for biochar production. Mean residence time of biochar depends on feedstock material, pyrolytic method used and the substrate where it is applied [8,24,25]. Fidel et al. [26] reported that biochar has the potential to improve the soil inorganic carbon by 0.023–0.045 mg C kg⁻¹ and organic carbon by 0.001–0.0069 mg C kg⁻¹. According to a study conducted by Ghosh and Maiti, [27], *Lantana camara* biochar lowered mine spoil CO₂ flux to 3% (2.60 µmol CO₂ m⁻² s⁻¹) and 2% (2.85 µmol CO₂ m⁻² s⁻¹) in comparison to control (4.92 µmol CO₂ m⁻² s⁻¹). Biochar acts as an amendment and improves physico-chemical, biological and nutritive soil properties [25,28]. An enriched soil supports the growth of vegetation which can facilitate ecosystem development and promote carbon sequestration in vegetation and soil. Thus, it is imperative to investigate the link between the intrinsic characteristics of biochar and mine spoil restoration.

The excessive growth of invasive weeds in RMS during the plantation stage of reclamation causes the problem of allelopathy [28]. These weeds are usually uprooted and left to decompose which adds to the atmospheric CO₂ pool. During the dry tropical summers, they act as fuel and cause even bigger problems of mine fire. One such weed growing abundantly in RMS is *Calotropis procera* (Aiton) W.T.Aiton (family: Apocynaceae). *C. procera* is a hardy shrub with an average height of 2 m and covered with a fissured corky bark which is high in cellulose and lignin. This can be a potential feedstock for biochar production and mine reclamation. Only a few studies have reported on biochar based carbon sequestration in a RMS, and the available data are from laboratory or greenhouse scale experiments [11,27]. The present study was conducted in an 8 year old RMS and the carbon sequestered was calculated in this RMS. A 6 month biochar based field experiment was also conducted to study the effect of biochar as an amendment for reclamation of mine spoil. The study aims to understand how coal mine reclamation along with biochar application can help in achieving UN SDG 13 and 15. Thus, the objectives of the study are: (i) assessment of carbon sequestration in an 8 year old RMS by vegetation, litter and soil carbon stock, (ii) application of *C. procera* biochar in the RMS in a 6 month field based study, (iii) fractionation of carbon in RMS, biochar amended RMS, reserved forest (RF) and unreclaimed mine spoil (UMS) (iv) calculation of total CO₂ sequestration in each system.

2. Materials and Methods

2.1. Site Description

The study area is located in Damoda colliery, Jharia Coalfield, situated in the Dhanbad district of Jharkhand, India ($23^{\circ} 39' - 23^{\circ} 48' \text{ N}$; $86^{\circ} 11' - 86^{\circ} 27' \text{ E}$). The site map of the study area is presented in Figure 1a,b. Damoda eco-restoration site is an 8 year old back-filled dump site of 4 ha area. The geology of the mine spoil consisted of sandstone, carbonaceous shale, intermixed shale and sandstone, Jhama (heat affected coal) with micaperidotite, subsoil and coal. The area experiences extreme weather conditions with summer temperature of 42 to 46 $^{\circ}\text{C}$ and winter temperature of 22 to 5 $^{\circ}\text{C}$ and received 1900 mm rainfall in the year of the study (2019). Jharia Coalfield is located in a dry tropical region and experiences three main seasons: summer, monsoon and winter. The carbon sequestration study was conducted in the February 2019, 6 months prior to which biochar was incubated in field conditions.

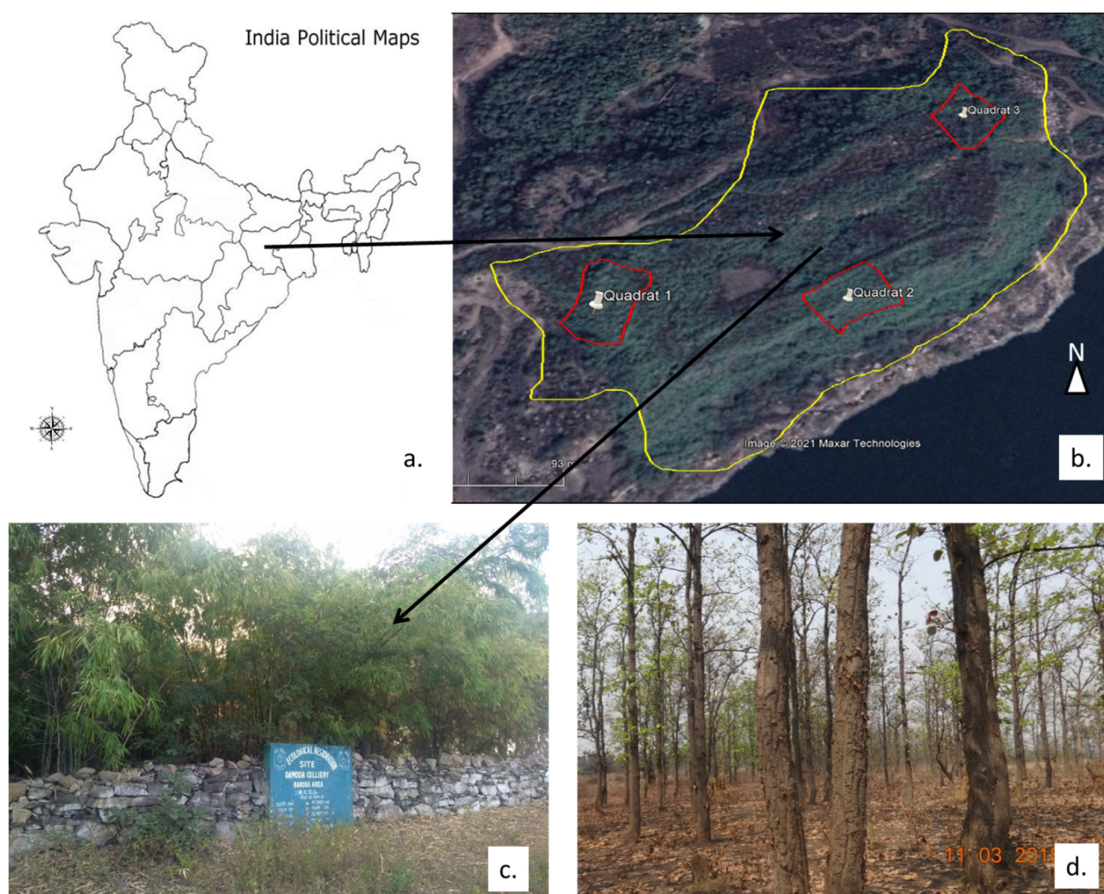


Figure 1. (a) Map of India, showing the Jharkhand state (b) Location map of Damoda eco-restoration site showing the 3 quadrats in the sampling area (c) Outer view of Damoda eco-restoration site showing dense bamboo clumps and stone boundary (d) Reserve forest sampling site.

The eco-restored mine dump has a history of shovel–dumper based mining activity. In 2011, plantation of hardy and multipurpose tree saplings was carried out in pits of dimension 30 cm \times 30 cm \times 30 cm. Grass seeds such as *Pennisetum pedicellatum* Trin. were also spread, which act as pioneer species and develop understory vegetation. Afforested trees such as *A. lebbeck*, *D. sissoo* and *Bambusa arundinacea* (L.) Voss were dominant species with sparse growth of plants such as *Azadirachta indica* A. Juss., *Bauhinia variegata* (L.) Benth., *Melia azedarach* L., *Psidium guajava* L., *Syzygium cumini* (L.) Skeels., *Terminalia arjuna* (Roxb.) Wight & Arn, and *Zizyphus mauritiana* Lam. Figure 1c shows the Damoda eco-

restoration site with stone boundary. RF area near the mining area was used as a positive reference site while UMS was used as a negative reference for the study. The most dominant trees in the RF site were *D. sissoo*, *A. lebbeck*, *Butea Monosperma* (Lam.) Taub. and *Shorea robusta* Roth (Figure 1d). UMS was coarser with rock debris, soils and subsoil materials. Since UMS was not revegetated, tree species were absent.

2.2. Biochar Production, Characterization and Field Incubation

C. procera growing in the RMS was collected in bulk, sun-dried, grinded and used for biochar production. Feedstock was pyrolysed in a muffle furnace at 450 °C for 60 min. Biochar characterization was carried out using the methods given in Ghosh et al. [11] and Ghosh and Maiti, [27]. The biochar field experiment was conducted as a completely randomized design in a 2 × 3 factorial scheme, each with 50 cm × 50 cm plots with two biochar application rate of 30 t ha⁻¹ (BC₃₀) and 60 t ha⁻¹ (BC₆₀), and each with three replications (Figure 2a–c). The carbon sequestration study was done with a 6 months incubation period in natural field conditions.



Figure 2. (a) Biochar being applied in RMS, (b) Plots showing biochar application (50 cm × 50 cm × 10 cm), (c) Ecological restoration project site of Damoda, showing boundary wall and sign board,

(d) Soil sampling being done by a soil corer (80 cm × 20 cm), (e,f) Collection of litter from a metal quadrat (50 cm × 50 cm).

2.3. Soil Sampling

Soil samples were collected from the rhizospheric region (0–15 cm) of different tree species, 8 samples collected from each quadrat with a metallic corer (80 cm × 20 cm) after removing the litter (Figure 2d). A total of 24 samples (8 samples × 3 quadrats) were collected from each RMS and RF. In the biochar incubation sites, 4 samples were collected from each plot making the total number of samples 24 (2 application level × 3 replicates × 4 samples per plot). Samples were placed in zip-lock packets and brought back to the laboratory for physico-chemical analysis. Samples were air-dried inside the laboratory for a week at room temperature.

2.4. Plant Biodiversity and Vegetation Analysis

Vegetation carbon stock was only analysed for RMS, RF and UMS, and no observable changes were observed with 6-month biochar application on the tree stocks, hence this data were not included. Three random quadrats of size 10 m × 10 m, covering total area of 300 m² were laid down for relative density [3]. Details on the density of the plants, IVI values and total number of species in RMS are provided in the Supplementary Materials. The density of species present in each site was expressed as number of individual species present per hectare area [3].

$$\text{Relative density (\%)} = \frac{\text{Number of individual plant species}}{\text{Total number of species in a quadrat}} \times 100 \quad (1)$$

Circumference of large tree species in the quadrats were measured at 1.37 m for the measurement of diameter at breast height (DBH), and the height of the tree was measured with a Distometer (Bosch GLM 40, India), while smaller vegetation (<3 m height) was measured using a digital Vernier caliper (Precise®, India). Specific gravity of the wood was measured by water displacement method. The aboveground biomass (AGB) was estimated by the regression model developed by Chave et al. [29], which showed the best-fit for tropical forests. The model estimated tree AGB by the following equation:

$$\text{AGB} = 0.0673 \times (\rho D^2 H)^{0.976} \quad (2)$$

where, AGB = above ground biomass (kg), ρ = wood specific gravity (g cm⁻³), D = DBH (cm) and H = tree height (cm).

Root biomass (RB) was calculated by multiplying AGB by a factor of 2.25 [30]:

$$\text{RB (Mg ha}^{-1}\text{)} = \text{AGB (Mg ha}^{-1}\text{)} \times 2.25 \quad (3)$$

Tree carbon stock was calculated multiplying a factor of 0.5 by total tree biomass [31].

$$\text{Tree C stock (Mg ha}^{-1}\text{)} = \text{Total tree biomass (Mg ha}^{-1}\text{)} \times 0.5 \quad (4)$$

The CO₂ sequestered by the plantation stock is calculated by relation given by IPCC [30].

$$\text{CO}_2 \text{ sequestered (Mg ha}^{-1}\text{)} = \text{Tree C stock (Mg ha}^{-1}\text{)} \times 3.67 \quad (5)$$

The AGB of bamboo clumps were calculated by the allometric relationship given by Nath et al. [32] and Mazumder et al. [33]. This equation was primarily developed to establish a relationship between culm height, density and AGB in thick walled bamboo. The equation is as follows:

$$\text{AGB} = 7.5 \times (D^2 H)^{0.91} \quad (6)$$

where H is total height of the bamboo culm, and D is DBH of the bamboo culm. 47% of the total biomass stock was considered as total carbon stock [32].

2.5. Herbaceous Biomass and Litter Analysis

The herbaceous biomass and litter present in the respective quadrats were measured by placing three litter traps (50 cm × 50 cm) under the tree canopy per quadrat [34] as shown in Figure 2e,f. The collected biomass was then dried in a hot air oven at 65 °C for 48 h. The dry weight of the litter obtained was converted to kg m⁻² by dividing it by quadrat area (50 cm × 50 cm) and then converting to kg ha⁻¹. Litter was assumed to have 40% carbon; hence C stock was calculated by a conversion factor of 0.4.

2.6. Soil Characterization

2.6.1. Soil Carbon Fractionation

Soil fractionation for the determination of inorganic, biogenic (labile and stable) and coal carbon present in the mine spoil was determined by the sequence of steps given by Ussiri and Lal. [10] The steps followed for sequential extraction of different forms of soil organic carbon, coal carbon and inorganic carbon in RMS, BC₃₀, BC₆₀ and RF are given in Figure 3.

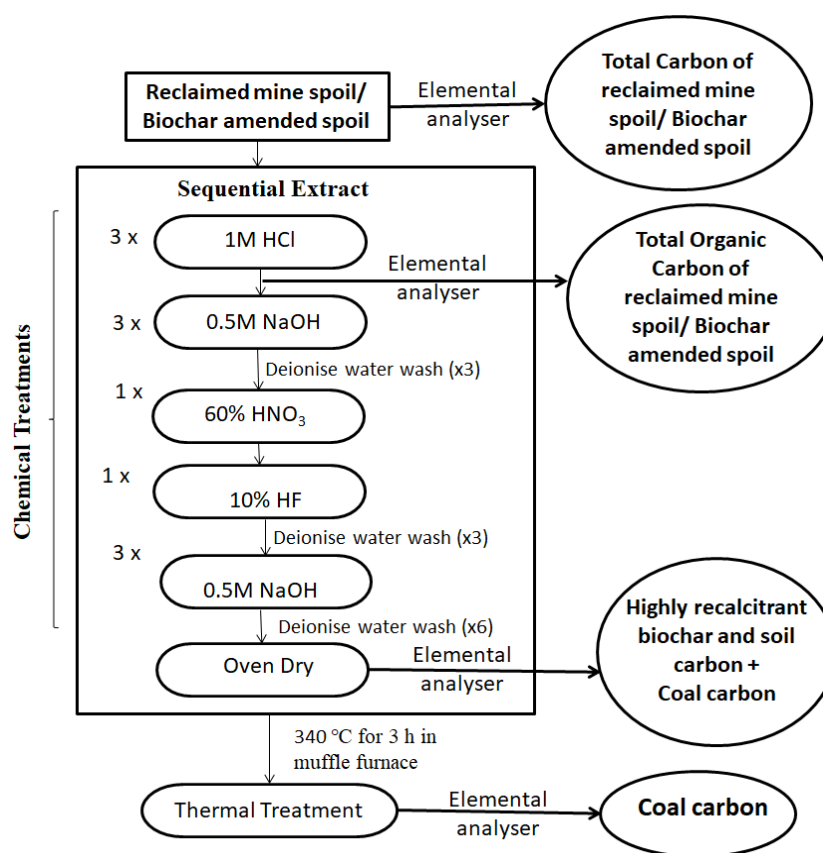


Figure 3. Sequential methods for the determination of total carbon, inorganic carbon and biogenic carbon pools in RMS, BC₃₀, BC₆₀ and RF (n x indicated the number of times the step was repeated).

2.6.2. Soil Physico-Chemical Properties

The soil samples were air dried and sieved by a 2-mm sieve to remove the coarse fraction from the fine earth fraction (< 2 mm). pH and EC were determined in a soil and water slurry (spoil: water, 1:2.5, w/v) by a multiparameter probe (HI-2020, Hanna Instruments, India). Cation exchange capacity (CEC) was calculated by the ammonium acetate

extraction method [35]. Available-N was determined by a Kjeldahl distillation unit (KJEL-ODIST-EAS VA, Pelican Equipment Inc. India). Available-P was extracted by NaHCO_3 (pH 8.5) and measured by a UV-VIS Spectrophotometer (Shimadzu Corporation, Japan) [36]. Available-K was calculated by 1 N ammonium acetate method by using flame photometer [5]. The C-stock is often underestimated due to the coarse fraction in mine soils, hence only soil fraction (<2-mm particle size) was considered for bulk density calculation. The bulk density was corrected by the equation given by Ahirwal et al. [6]:

$$\text{Corrected Bulk Density (Mg m}^{-3}\text{)} = \frac{\text{Sample weight (Mg)} \times \text{Fine earth fraction (\%)}}{\text{Volume of corer (m}^3\text{)} \times 100} \quad (7)$$

Soil organic carbon (SOC) of the study sites were calculated by the relation [6]:

$$\text{SOC stock (Mg ha}^{-1}\text{)} = \frac{\text{Biogenic carbon pool(\%)} \times \text{BD (Mg m}^{-3}\text{)} \times T \text{ (m)} \times 10^4 \text{ (m}^2\text{ha}^{-1}\text{)}}{100} \quad (8)$$

where SOC = Soil organic carbon; BD = corrected bulk density; and T = depth of the soil layer.

2.7. Carbon Sequestration Study

The total C sequestration pool of an ecosystem is calculated by adding the C-stock associated with (i) AGB and RB, (ii) understory vegetation and litter layer, and (iii) SOC stock. Carbon is accumulated in vegetative parts such as leaf, twigs, and logs, live and dead roots, and soil organic matter. The biomass carbon pool varies from plant to plant and also by age of vegetation. Thus, the total ecosystem carbon pool can be assessed by adding (i) AGB and RB carbon-pool, (ii) understory and litter C stock and (iii) biogenic carbon stock at (0 to 15 cm). To determine the Carbon sequestration rate ($\text{Mg C ha}^{-1} \text{ year}^{-1}$), total C stock (Mg C ha^{-1}) was divided with by age of reclamation.

2.8. Statistics

One-way ANOVA was used to compare the means of data obtained from RMS, BC_{30} , BC_{60} and RF. Post hoc Duncan's multiple range tests at $p < 0.05$ significance level was used to test the significant difference in the C-stock in each level of analysis. SPSS 23 was used for statistical studies, and software such as MS-EXCEL and ORIGIN Pro-8 was used for graphical representation.

3. Results and Discussions

3.1. Biochar Characteristics

The general characteristics *C. procera* biochar are presented in Table 1. Biochar yield obtained from *C. procera* feedstock was 51.87%. Biochar obtained was alkaline in nature with a pH of 7.75 and EC of 4.7 mS cm^{-1} . The total elemental C, H and N were 68.25%, 35.39% and 13.62% respectively. The C/N ratio of 5.01 indicates that the biochar is rich in labile carbon, providing substrate for microbial action in the mine spoil, while H/C ratio of 0.51 for *C. procera* biochar represents its high degree of aromatization. *C. procera* has a high organic carbon content of 42.24%, porosity of 78% and low bulk density of 0.25 g cm^{-3} .

Table 1. Physio-chemical properties of *C. procera* biochar ($n = 5$, mean \pm standard deviation).

Characteristics	Values
Yield (%)	51.87 \pm 2.27
pH _{Water} (1:5; w/v)	7.75 \pm 1.62
EC _{Water} (1:5; w/v) (mScm ⁻¹)	4.70 \pm 0.12
C (%)	68.25 \pm 4.58
H (%)	35.39 \pm 5.22
N (%)	13.62 \pm 2.40
H/C	0.51 \pm 0.22
C/N	5.01 \pm 1.28
OC (%)	42.24 \pm 0.89
Porosity (%)	78 \pm 4.00
Bulk density (g cm ⁻³)	0.25 \pm 0.01

The high surface area and the porous morphology of the biochar surface can be seen in the FE-SEM image of *C. procera* biochar given in Figure 4a,b. The porous structure provides an enlarged surface area and substrate for microbial action. Several spectral peaks representing various functional groups were observed on the *C. procera* biochar surface (Figure 4c). At transmittance of 3391 cm⁻¹ an O–H bond is prominent due to the breaking of hydrogen bonded hydroxyl groups. Other bonds such as –CH₃ (2924 cm⁻¹), –CH₂ (2870 cm⁻¹), C = O (1600–1700 cm⁻¹), due to cellulose of the feedstock, are also present. The peaks at 500–600 cm⁻¹ represent the aromatic carbons in the biochar surface.

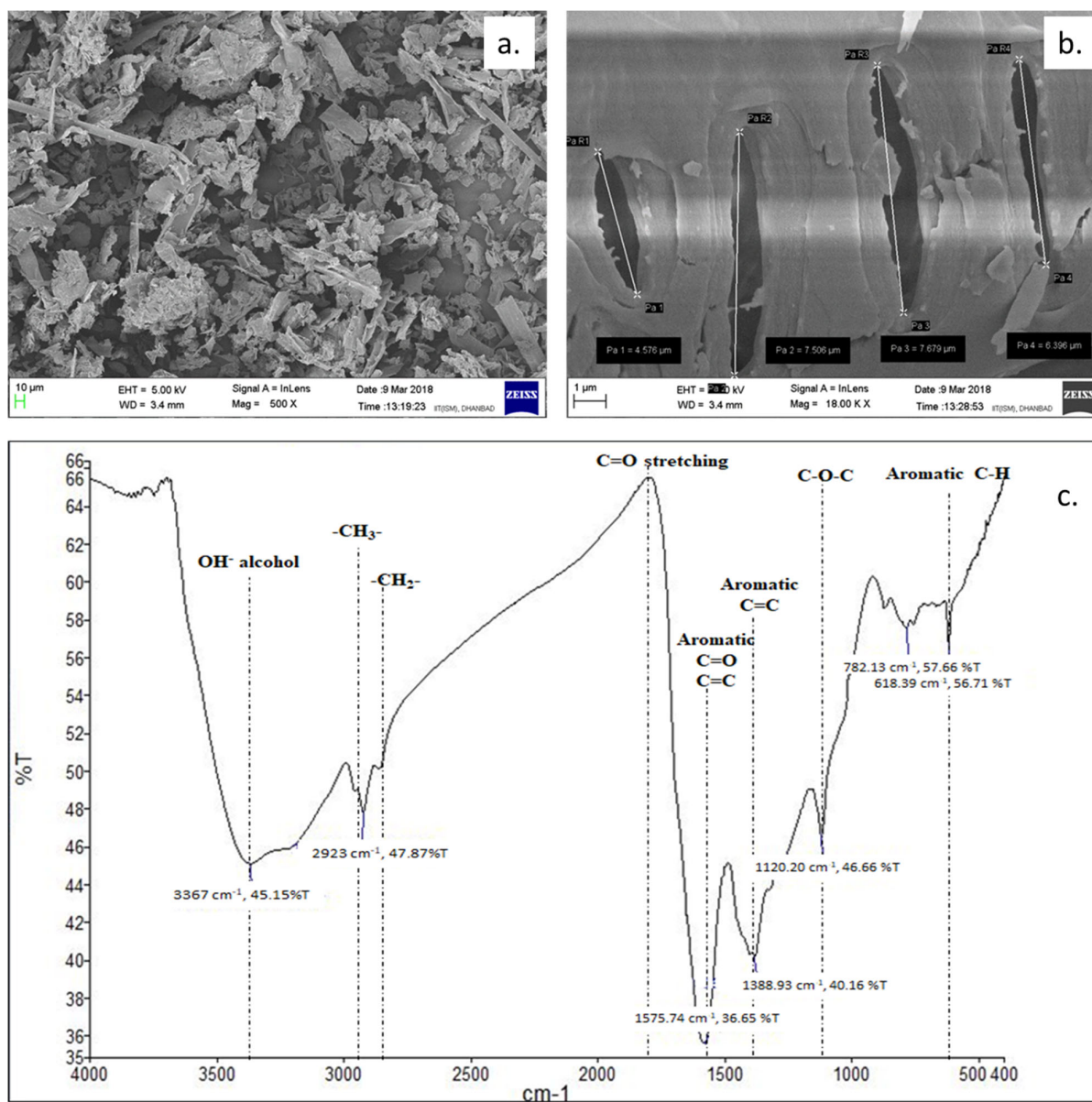


Figure 4. (a) FE-SEM image of *C. procera* biochar at 500 \times magnification, (b) FE-SEM image of *C. procera* biochar at 1800 \times magnification showing the pore sizes, (c) FTIR spectra of *C. procera* biochar showing the surface functional groups.

3.2. Plant Biodiversity and Vegetation Analysis

B. arundinacea clump density was 4033 clumps ha^{-1} , whereas the tree density was 3233 trees ha^{-1} . Relative distribution of the species in the reclaimed site is shown in Figure 5. *B. arundinacea* clumps are most abundant (56%), followed by *Albizia* spp. (18%), *D. sissoo* (10.5%), and *Z. mauritiana* (5%). Das and Maiti, [37] reported that the same reclaimed mine spoil at 4 years old had a bamboo clumps density of 3033 clumps ha^{-1} , whereas the tree density was 2500 trees ha^{-1} .

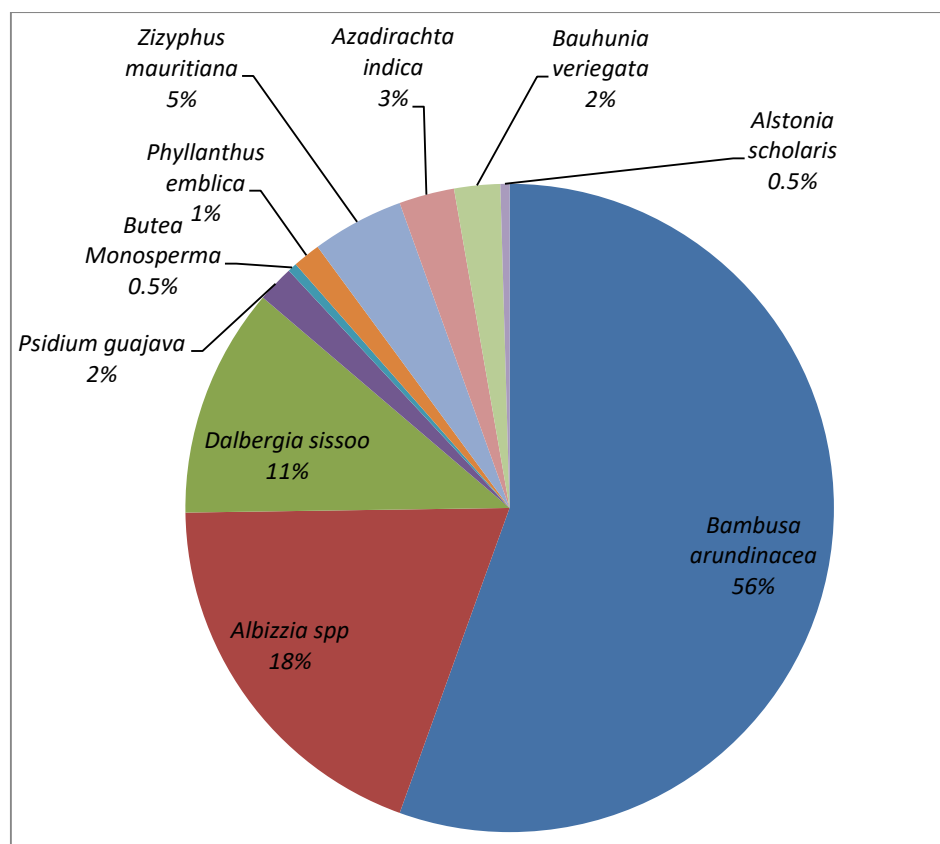


Figure 5. Relative density of each species in the RMS showing the percentage of each species in the study area.

3.3. Estimation of Biomass Carbon Stock

Plant biomass and C pool associated with RMS and RF are summarized in Table 2. In RMS, *Albizzia spp.* ($4.34 \text{ Mg C ha}^{-1}$) had the highest total tree carbon stock, followed by *B. arundinacea* ($2.61 \text{ Mg C ha}^{-1}$) and *D. sissoo* ($2.91 \text{ Mg C ha}^{-1}$). The total carbon stock from the plant biomass was $12.59 \text{ Mg C ha}^{-1}$ and CO_2 sequestered was 48.17 Mg ha^{-1} . Tree biomass of the RF was three times ($30.63 \text{ Mg C ha}^{-1}$) higher than the RMS ($12.59 \text{ Mg C ha}^{-1}$) and UMS ($3.52 \text{ Mg C ha}^{-1}$). Čížková et al. [38] reported $1.6 \text{ t ha}^{-1} \text{ y}^{-1}$ potential for carbon sequestration in reclaimed grasslands in a reclaimed lignite mine. Ahirwal et al. [39] reported the effect of fast-growing trees on soil properties and the ecosystem carbon pool after eight years of afforestation. The study reported greater carbon storage in *D. sissoo* (39 Mg C ha^{-1}) compared to *A. lebbeck* (34 Mg C ha^{-1}) and *A. procera* (26 Mg C ha^{-1}). In a 16 year reclaimed coal mine site, Ahirwal and Maiti, [40] reported that the tree carbon stock was 75% of the reference forest site, plantation of multipurpose tree species improved mine spoil fertility, facilitates natural growth of indigenous tree species. Due to the plant soil-interaction in a coal mine spoil, the roots of the re-vegetated plants alter soil structure and function [8]. Although carbon stock associated with RMS was less than that of RF, yet the growth of native species proves that proper reclamation technology can influence ecosystem development. Thus, increase in C-stock proves that plantation is a successful means of mine reclamation which certainly helps in achieving SDG 13 and 15.

Table 2. Total biomass, tree carbon stock and CO₂ sequestered in various coal mine spoils of the world and their values in the current study (RMS: reclaimed mine spoil and RF: reserve forest).

Reclamation Type	Location	Age	Total Biomass (Mg C ha ⁻¹)	Tree Carbon Stock (Mg C ha ⁻¹)	CO ₂ Sequestered * (Mg ha ⁻¹)	References
<i>Albizia lebbeck</i>	Singrauli, India	4–5	17.28	8.64	31.70	[41]
<i>Albizia procera</i>			8.48	4.24	15.56	
<i>Tectona grandis</i>			7.34	3.67	12.36	
<i>Dendrocalamus strictus</i>			15.02	7.51	27.56	
Mixed Plantation	Singrauli, India	10	15.64	13.68	50.20	[18]
Reclaimed dump	Jharia, India	5	18.07	9.03	33.14	[42]
Reference forest			55.25	27.63	101.40	
<i>Albizia lebbeck</i>	Jharia, India	8	73.66	34.62	127.05	[39]
<i>Albizia procera</i>			55.76	10.89	39.96	
<i>Dalbergia sisso</i>			83.28	26.21	96.19	
Natural forest			250.28	117.63	431.702	
RMS	Damoda eco-restoration,	8	25.18	12.59	46.17	Present Study
RF			61.26	30.63	112.41	
UMS	Jharia, India		7.05	3.52	12.93	

* CO₂ sequestered (Mg C ha⁻¹) = Tree carbon stock (Mg C ha⁻¹) × 44/12.

3.4. Herbaceous Biomass and Litter Analysis C-Stock

Litter consists of twigs, plant debris, foliage and branches which possess high nutrient content. It is one of the most important sources of organic matter in the mine spoils, and litter decomposition contributes to nutrients recycling and improvement in soil fertility [36]. During reclamation of a degraded mine spoil, increase in litter C improves the SOC of the ecosystem and can be an indicator of restoration success. Litter and understory carbon stock in RMS, BC₃₀, BC₆₀, RF and UMS are presented in Figure 6. The total carbon stock in RMS was 1.24 Mg ha⁻¹, while higher values of 1.64 Mg ha⁻¹ and 1.73 Mg ha⁻¹ were observed for BC₃₀ and BC₆₀ respectively. Biochar application had carbon stock comparable to the RF (1.79 Mg ha⁻¹) and was significantly higher ($p < 0.05$) than UMS (0.5 Mg ha⁻¹).

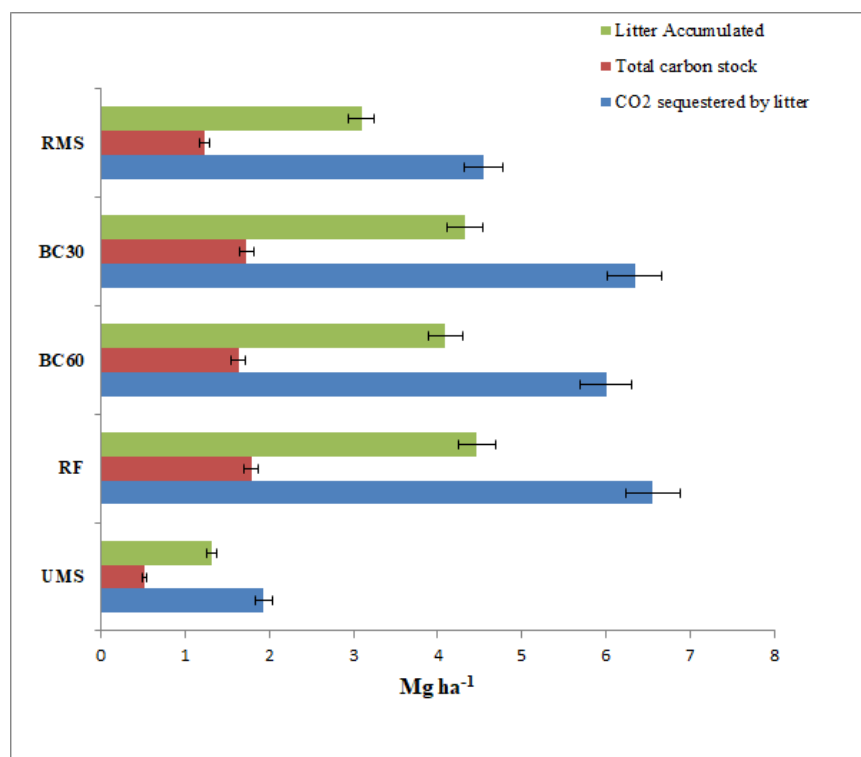


Figure 6. Litter and understory carbon stock in reclaimed mine spoil (RMS), biochar treatment at 30 t ha⁻¹ (BC₃₀) and 60 t ha⁻¹ (BC₆₀), reserved forest (RF) and unreclaimed mine spoil (UMS) ($n = 24$).

3.5. Mine Spoil Properties

3.5.1. Inorganic, Biogenic and Coal Carbon Estimation

Carbon fractions in RMS, BC₃₀, BC₆₀, RF and UMS are presented in Figure 7a. The total soil carbon at BC₃₀ (36.3 g C kg⁻¹) and BC₆₀ (40 g C kg⁻¹) was found to be significantly high ($p < 0.05$) compared to RMS (21 g C kg⁻¹), comparable to RF (33 g C kg⁻¹), and low in UMS (12 g C kg⁻¹). Although there was no significant difference between the soil inorganic carbon of RMS (1.9 g C kg⁻¹), RF (2 g C kg⁻¹) and UMS (0.7 g C kg⁻¹), the inorganic fraction was found to increase to 3.5 g C kg⁻¹ and 4.5 g C kg⁻¹ at BC₃₀ and BC₆₀, respectively. Average coal carbon was higher in RMS, BC₃₀, BC₆₀ and UMS compared to RF. A greenhouse experiment conducted by Rodríguez-Vila et al. [43] on copper mine spoils reported a range of 20–207 g C kg⁻¹ for total soil carbon and 3–27 g C kg⁻¹ for inorganic carbon by biochar application rate of 20–100%.

Biogenic carbon fraction is a complex pool which can be broadly divided into labile and recalcitrant carbon pool. Labile pool has a residence time of years to a few decades while the recalcitrant carbon pool can remain in the soil for hundreds to thousands of years [37]. Figure 7b shows the labile and the recalcitrant carbon present in RMS, BC₃₀, BC₆₀ and RF. Labile fraction was found to be 56% and 51% of biogenic carbon pool for RMS and RF, respectively. The labile carbon trend was of the order UMS > RMS > RF > BC₃₀ > BC₆₀. Recalcitrant fraction was found to be 44% and 49% of the biogenic carbon pool for BC₃₀ and BC₆₀, respectively. The recalcitrant carbon trend was of the order BC₆₀ > BC₃₀ > RF > RMS > UMS. Labile C pools can have a large impact on soil–biochar interactions in the short term (~6 months), whereas recalcitrant carbon pool is influential in soil properties and soil function for a longer period of time [24,26]. Sub-fractions of soil organic carbon are indicators of soil fertility and are instrumental in understanding the influence of a management practice. In a study conducted on a 10 year old reclaimed coal mine spoil, Das and Maiti. [44] reported that the biogenic C constituted 45–66% of total soil carbon in RMS. Fidel et al. [26] reported that biochar improves the soil inorganic carbon by 0.023–

0.045 mg C kg⁻¹ and organic carbon by 0.001–0.0069 mg C kg⁻¹ in eroded soil. The study also reported that labile biochar pools are stabilized by the recalcitrant C pool. Thus, because of the higher recalcitrant fraction, biochar application will play an instrumental role in increasing the C-stock and help achieve the UN SDG 13 and 15.

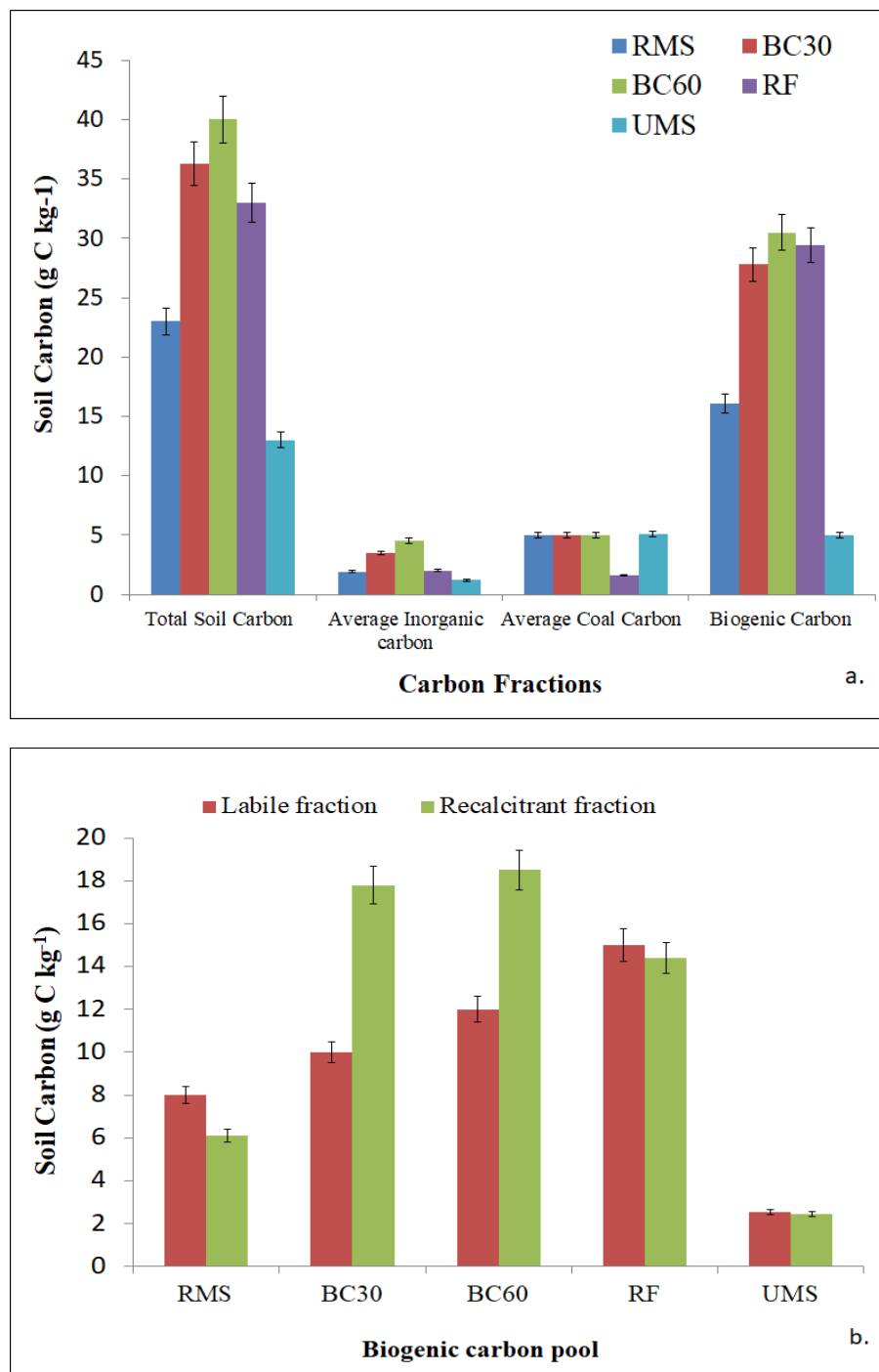


Figure 7. (a) Comparison of different carbon fractions in reclaimed mine spoil (RMS), biochar treatment at 30 t ha⁻¹ (BC₃₀) and 60 t ha⁻¹ (BC₆₀), reserved forest (RF) and unreclaimed mine spoil (UMS). (b) Comparison of labile and recalcitrant carbon fractions in RMS, BC₃₀, BC₆₀, RF and UMS.

3.5.2. Other Physio-Chemical Properties

RMS, BC₃₀ and BC₆₀, RF and UMS properties are summarized in Table 3. Soil fraction was significantly ($p < 0.05$) high in the reference forest compared to the 8 year old RMS and UMS, biochar application had no effect in the soil fractions. pH of RMS was neutral, biochar application resulted in an alkaline mine spoil, while it was slightly acidic in the RF and UMS. EC of the RMS was 0.16 dS cm⁻¹ compared to 0.11 dS cm⁻¹ in RF soil, 0.17 dS cm⁻¹ in UMS, 0.09 dS cm⁻¹ in BC₃₀ and 0.1 dS cm⁻¹ in BC₆₀. CEC was higher in RF (13.1 C mol kg⁻¹) compared to RMS (8.22 C mol kg⁻¹). Moisture content in RF was 26% higher than of RMS, while BC₆₀ and BC₃₀ improved the moisture content by 33% and 55% respectively ($p < 0.05$). Available N and P showed significant difference ($p < 0.05$) in the RMS and RF; available N and P in RF were 26% and 47% higher, respectively. The exchangeable potassium was also found to be higher in RF (55.22 mg kg⁻¹) compared to the 8 year old RMS (102.1 mg kg⁻¹). BC₃₀ and BC₆₀ improved the NPK values in the mine spoil significantly ($p < 0.05$) which helps in vegetation growth. The corrected bulk density was higher in the RF site (1.34 Mg m⁻³) compared to the RMS (0.71 Mg m⁻³), BC₃₀ (0.65 Mg m⁻³) and BC₆₀ (0.63 Mg m⁻³). Ghosh et al. [11] reported an increase in organic carbon by threefold, CEC by twofold, with a decrease in bulk density to half, by *Lantana* biochar application in a coal mine spoil. Thus, biochar application can effectively improve the physio-chemical properties of the mine spoil which will help accelerate the process of soil formation in a RMS and increase C-stock to near RF level. The application of biochar improves the soil physico-chemical properties and increases the carbon stock in the soil. This ameliorated mine spoil will support plant growth and help in ecosystem development. This in the long run will help in achieving the UN SDGs 13 and 15.

Table 3. Characteristics of RMS (reclaimed mine soil), BC₃₀ (biochar @ 30 t ha⁻¹), BC₆₀ (biochar @ 60 t ha⁻¹), RF (reference forest) soil and unreclaimed mine spoil (UMS) ($n = 24$, mean \pm Standard deviation, within each row, values with same letter are not significantly different, $p < 0.05$ with Duncan's multiple range test).

Soil Parameters	RMS	BC ₃₀	BC ₆₀	RF	UMS
Soil Fraction (< 2 mm size)%	63.10 \pm 12.06 b	63.10 \pm 12.06 b	63.10 \pm 12.06 b	88.12 \pm 6.57 a	25.6 \pm 3.4 c
Non- Soil Fraction (> 2 mm size)%	36.89 \pm 12.04 b	36.89 \pm 12.04 b	36.89 \pm 12.04 b	11.88 \pm 3.20 c	74.4 \pm 3.3 a
pH(water, 1:2.5, w/v)	7.15 \pm 0.41 b	9.66 \pm 0.2 a	9.53 \pm 0.57 a	6.12 \pm 1.22 c	6.3 \pm 0.4 c
EC (water, 1:2.5, w/v) dS/m	0.16 \pm 0.27 a	0.09 \pm 0.3 c	0.1 \pm 0.56 b	0.11 \pm 0.08 b	0.17 \pm 0.06 ab
CEC (C mol kg ⁻¹)	8.22 \pm 1.55 c	13.25 \pm 4.22 b	18.39 \pm 0.38 a	13.1 \pm 0.27 b	5.22 \pm 1.54 d
Moisture Content (%)	5.37 \pm 5.35 d	8.14 \pm 0.12 b	10.77 \pm 0.28 a	7.29 \pm 2.18 c	4.26 \pm 2.6 d
Available-N (mg kg ⁻¹)	96 \pm 8.34 cd	102 \pm 5.87 b	105 \pm 6.27 b	130 \pm 5.22 a	58.72 \pm 4.2 d
Available-P (mg kg ⁻¹)	3.82 \pm 0.84 d	8.91 \pm 1.12 b	10.18 \pm 0.4 a	7.26 \pm 0.87 bc	3.24 \pm 1.3 d
Exchangeable K (mg kg ⁻¹)	55.22 \pm 3.57 c	423.3 \pm 35.11 a	456.66 \pm 7.4 a	102.1 \pm 5.22 b	30.56 \pm 3.2 d
Corrected bulk density (Mg m ⁻³)	0.71 \pm 0.51 b	0.65 \pm 0.58 c	0.63 \pm 0.91 c	0.91 \pm 0.85 ab	1.05 \pm 0.10 a
SOC (Mg ha ⁻¹)	31.33 \pm 0.75 c	41.29 \pm 1.22 b	45.7 \pm 1.89 b	72.11 \pm 5.22 a	12.6 \pm 0.32 d

3.6. Total C-Pool

The total C stock, CO₂ sequestered and rate of C sequestration in RMS, BC₃₀, BC₆₀, RF and UMS are presented in Table 4. The C stock of RF (72.11 Mg C ha⁻¹) was almost thrice the RMS (30.98 Mg C ha⁻¹) and 5 times the UMS (13.92 Mg C ha⁻¹). Application of biochar @ 30 t ha⁻¹ improved the C-stock of RMS by 33%, but was 42% lower than the RF. Similarly, biochar @ 60 t ha⁻¹ improved the C-stock of RMS by 47%, but was 36% lower than the RF C-stock. CO₂ sequestered had the sequence, RF (264.64 Mg C ha⁻¹) > BC₆₀ (168.22 Mg C ha⁻¹) > BC₃₀ (151.70 Mg C ha⁻¹) > RMS (113.69 Mg C ha⁻¹) > UMS (48.37 Mg C ha⁻¹). Mukhopadhyay and Masto [45] reported yard waste biochar improved the stable carbon pool in biochar amended mine spoil to 0.873 g CO₂-C kg⁻¹ compared to 0.03 g CO₂-C kg⁻¹ in mine spoil. Xu et al. [46] reported that the application of bamboo leaf biochar at 5 and 15 Mg ha⁻¹ application rate increased the ecosystem carbon stock by 1486.31% and 252.98%, respectively. This increase could be due to the increase in vegetative cover with time. Plant

growth can play an important role in decomposition and humus layer formation, which later turned into soil organic matter and increases the organic carbon pool of the soil and provides nutrients to the reclaimed vegetation.

Application of biochar increased the C-stock of RMS by 33%, and 47% at application rates of 30 t ha⁻¹ and 60 t ha⁻¹, respectively. As mentioned earlier, recalcitrant fraction of this C-stock is 44% and 49% of the biogenic carbon pool for BC₃₀ and BC₆₀, respectively. Thus, it can be concluded that the increase in the C-pool in the 6 months of the study was due to the labile fraction. The remaining 44% and 49% will remain in the form of recalcitrant carbon in the RMS, and thus, will aid in achieving the climate action goals of the SDG.

Table 4. Effect of biochar application on carbon stocks of different landforms. Comparison of total C stock, CO₂ sequestered, Rate of C accumulation in 8 year old RMS (reclaimed mine spoil), BC₃₀ (biochar @ 30 t ha⁻¹), BC₆₀ (biochar @ 60 t ha⁻¹), RF (reserve forest) and unreclaimed mine spoil (UMS).

Land use	Country/Location	Biochar Feedstock	Results	References	
Sub-urban red soil	Hangzhou, China	Oak wood, bamboo	-Lability index increased by 4 and 6%, respectively, -The carbon management index (CMI) increased by 50 to 286%.	[24]	
Agricultural Soil	Pottawattamie County, USA	Corn stover	Increase in soil inorganic carbon by 0.023–0.045 mg C kg ⁻¹ and organic carbon by 0.001–0.0069 mg C kg ⁻¹ .	[26]	
Moso bamboo forest	Zhejiang, China	Bamboo leaf	5 and 15 Mg ha ⁻¹ increased the ecosystem carbon stock by 1486.31% and 252.98%, respectively.	[46]	
Agricultural soil	Atlantic, USA	Wood	C stocks nearly twice (14.07 Mg soil C ha ⁻¹) the amount of C added with biochar 6 years earlier (7.25 Mg biochar C ha ⁻¹)	[47]	
Fresh Coal Mine spoil	Jharia, India	Yard waste	Stable carbon pool in biochar amended mine spoil was 0.873 g CO ₂ –C kg ⁻¹ compared to 0.03 g CO ₂ –C kg ⁻¹ in mine spoil	[45]	
Copper Mine spoil	Touro, Spain	Holm oak wood	20–207 g C kg ⁻¹ for total soil carbon and 3–27 g C kg ⁻¹ for inorganic carbon by biochar application rate of 20–100%	[43]	
			C stock (Mg C ha ⁻¹)	CO ₂ sequestered (Mg ha ⁻¹)	
RMS	Damoda, Eco-restoration,	<i>Calotropis procera</i>	30.98 ± 1.25	113.69 ± 3.5	Present Study
BC30			41.34 ± 1.3	151.70 ± 5.31	
BC60	Jharia, India		45.84 ± 1.5	168.22 ± 4.25	
RF			72.11 ± 3.2	264.64 ± 5.65	
UMS			13.18 ± 0.87	48.37± 1.27	

4. Future Recommendations

Through the course of this study, future goals and recommendations for continuing research in this field are as follows:

- A biochar based chrono-sequence study in the RMS to study the trends in carbon stock for a prolonged period of time.
- Field based long term studies are needed to understand the behaviour of carbon that is fixed in the soil by biochar application. Studies need to be done to ensure that the carbon in the biochar is fixed in soil for a long period of time and not emitted into the atmosphere.
- Effect of biochar application on the existing humus of the RMS.
- Conducting life cycle assessment (LCA) to confirm that net soil–ecosystem C pools is increased by biochar application.

- (v) Environmental cost benefit by biochar application should be carried out for the applicability of biochar.
- (vi) Developing techniques for the large-scale production of biochar in the field itself will help reduce the transportation cost.

5. Conclusions

Plantation of hardy species in coal mine spoil restores the derelict ecosystem which promotes natural colonization of indigenous species. In the current study, 8 year old RMS effectively increased the biomass, litter and biogenic carbon stock in the soil. The rate of C accumulation for 8 year old RMS was calculated to be 3.92 Mg C ha⁻¹ year⁻¹. Application of *C. procera* biochar in the RMS improved the soil physio-chemical properties. The inorganic and biogenic carbon pool, especially the recalcitrant pool, was improved by biochar application, suggesting that biochar can be an effective mode of enhanced carbon fixation in the spoil, along with plantation activities. A mere 6 month application period increased the C-stock by 36–42%, thus its recalcitrant carbon content can be fixed in the mine spoil for a longer period of time. This proves that biochar has tremendous potential in fixing carbon, along with forestry based reclamation of coal mine spoil. Thus, carbon stock increases with age of reclamation, and biochar application can increase the carbon stock close to reference forest site level. As the biochar–plantation synergy can both sequester carbon and also promote biodiversity, it can be an effective tool for achieving United Nations SDG 13 and 15.

Supplementary Materials: The following are available online at www.mdpi.com/article/10.3390/land10111112/s1, Table S1. List of species recorded in quadrats of RMS sites showing some biodiversity parameters.

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