

# Utilization of Agro-Industrial Wastes for Restoration of Degraded Land, Mitigation of Heavy Metals Pollution and Greenhouse Gases Emissions from Selected Industrial Areas of Bangladesh

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## Abstract

Heavy metals are the most significant contaminants for environment arising mainly from industrial activities. Two field experiments were conducted to investigate the soil contamination level, potential remediation of heavy metals pollution, and mitigation of greenhouse gases emissions through interactive effects of rice husk biochar, vermicompost, azolla compost with cyanobacteria, tea waste biochar and oyster shell biochar amendments with reduced number of chemical fertilizers in contaminated fields adjacent to Industrial areas of Mymensingh district. The experimental findings revealed mean maximum concentrations of heavy metals 1.65 mg/kg, 49.5 mg/kg, and 1340 mg/kg for Cd, Pb, and Mn in contaminated soils, which were decreased by Rice husk biochar with Oyster shell, Tea waste biochar with Oyster shell, and Vermicompost with Azolla Cyanobacteria dual cropping with rice. Higher contamination factors for Cd (CF 5.43 - 5.60), Pb (CF 2.28 - 2.64) and Mn (CF 1.45 - 1.71) than that of Cu (1.08 - 1.24) and Zn (0.58 - 0.73), indicates Cd, Pb and Mn were the main pollutants in the study areas. The maximum soil pollution load index (SPLI) value were estimated 1.70 - 1.80, and the maximum potential ecological risk index (ERI) value 184 - 188 and 176 - 185 were estimated for contaminated sites without any soil amendments. The decrease in pollution loads and improvement in ERI through Rice husk biochar, Tea waste biochar and Vermicompost amendments with Oyster shell may be due to the dissolution of metal pollutants. Among the amendments, Vermicompost with

silicate fertilization and Azolla Cyanobacterial dual cropping with rice increased rice yield by 10.0% - 13.8% and decreased cumulative methane (CH<sub>4</sub>) emissions by 9.0% - 13.0%; while rice husk biochar and tea waste biochar with silicate fertilizer amendments were found effective for reducing (27.0% - 36.0%) seasonal cumulative nitrous oxide (N<sub>2</sub>O) emissions compared to chemical fertilized field plot. The mean maximum GWP 5380 - 5575 kg CO<sub>2</sub> eq. ha<sup>-1</sup> estimated from a field plot without any organic amendments, was decreased by 14.5% - 15.7%, 11.5% - 14.5% and 7.5% - 12.5% with Vermicompost and silicate fertilization, Tea waste biochar and silicate fertilization and Vermicompost amendments with Azolla Cyanobacteria dual cropping with rice in wetland paddy field ecosystem. The SOC contents and CMI value increased significantly in the amended field soils at both locations under cropping pattern Rice-Sesbania-Rice.

### Keywords

Soil Restoration, Heavy Metals, Silicate, Vermicompost, Tea Waste Biochar, Oyster Shell, CMI, GWP

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## 1. Introduction

Agricultural ecosystem plays an important role in carbon sequestration. However, the rapid growth of industrialization and urbanization disrupts the agroecosystem through indiscriminate disposal of solid wastes, industrial effluents and other toxic wastes, which are posing threats to agricultural farming and the existence of biotic community. It has been reported that soil contamination level became severe in Bangladesh due to its growing industries without proper planning and their indiscriminate activities to the ecosystem [1]-[3]). Heavy metals released from industries are considered as pollutants due to their toxic effects on plants, animals and human beings. They interfere with physiological activities of plants such as photosynthesis, gaseous exchange and nutrients absorption, thereby causing a reduction in plant growth, dry matter accumulation and yield [4]. Rice (*Oryza sativa* L.) cultivation is a major agricultural land use in Bangladesh. The double-cropping rice production system mainly dominates in this country. Heavy metals pollution, particularly cadmium (Cd), lead (Pb), chromium (Cr), iron (Fe) and manganese (Mn) toxicity is a major issue for environmental degradation in Bangladesh. Mymensingh City Corporation including Bhaluka is an emerging industrial site, which has already become contaminated due to untreated industrial wastes disposal in the open field, surface drains and canals. The toxic heavy metals in soils around the industrial area may come from textile and dyeing industrial untreated waste disposal, garments manufacturing wastes, Pharmaceutical, Plastic and beverage company wastes, Lead battery factory waste, Ceramics and glass manufacturing wastes etc. As a consequence, agricultural crops and vegetables, fish species, including other aquatic animals, and human life are threatened in

this area due to environmental pollution. To assess multiple risks of toxic metals in soil, the pollution load index (PLI) and potential ecological risk index (PER) have been developed [5]. The PLI compares the metal concentrations with baseline values, which helps in assessing the enrichment of toxic metals in soil [6] [7].

Biochar as an adsorbent, having high porosity and surface area, may effectively remove heavy metals from waste in contaminated soil and water through interaction mechanisms such as physical adsorption, precipitation, ion exchange and electrostatic attraction [8]-[10]. It has been shown that plant-derived biochar has high efficiency in the binding of potentially metals such as Cu, Cd, Ni and Pb to form metal complexes with carboxylic and phenolic functional groups compared with biochar prepared from animal by-products such as dairy manure and poultry litter [11] [12]. Application of biochar may efficiently remove Cd and Pb from contaminated soil through precipitation, adsorption, or functional complexation [8]. Calcium, an essential macronutrient, can be used as an exogenous substance to alleviate Cd- and Pb-induced toxicity in plants. Ion exchange ( $\text{Ca}^{2+}$ ) is the dominant mechanism responsible for Cd and Pb removal by Ca-rich biochars. Natural oyster shells are biogenic materials composed of calcium carbonate (>95%) in association with an organic fraction (approximately 5%). Therefore, Oyster shell biochar (Ca rich) could be used for heavy metals immobilization and environmental restoration. The dissolution of  $\text{CaCO}_3$  from Oyster shell produces hydroxyl ion,  $\text{OH}^-$ , which increases the negative soil surface charge, thereby increasing Cd and Pb metal adsorption capacity [13]. Different long-term organic manures and inorganic chemical fertilizers especially N inputs have profound effects on soil properties and GHGs emissions. In addition, Silicon (Si) based fertilizer and soil-available silicon (Si) has been recognized as a vital component for agricultural production system, to reduce the toxicity of heavy metals [14], and mitigation of GHGs emissions [15]. In this regard, rice husk biochar (RHB), by-product of rice milling factory, mainly carbon (C), silicon (Si), potassium (K) and sulphur (S) enriched materials, obtained under high pyrolysis temperature ( $600^\circ\text{C}$ ) and oxygen limited condition, could be the potential organic manure to improve soil fertility, increase the mean residence time of soil organic C (SOC) content, reduce nutrients leaching and mitigate greenhouse gases emission [16] [17]. Rice cultivation is one of the major agricultural practices in Mymensingh Sadar and Bhaluka industrial areas, which has been decreasing due to soil contamination with pollutants and effluents discharged from the emerging Industries around Mymensingh region. In addition,  $\text{CO}_2$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  gases have been emitting from the contaminated water-logged paddy ecosystem, eventually enhancing the warming effects (GWPs). Azolla is a free-floating fern, recognized to modify the wetland paddy ecosystem through its symbiosis with nitrogen ( $\text{N}_2$ ) fixing Cyanobacteria (*Anabaena azollae*), mobilizing fixed phosphates and retarding  $\text{NH}_3$  volatilization during rice cultivation [18]-[20].  $\text{CH}_4$  emissions from the water logged paddy field could be controlled by modifying the paddy soil ecosystem through water management, or-

ganic and inorganic fertilizers amendments, addition of silicate fertilizer [15] and by accelerating the biological activity of Cyanobacteria *Azolla anabaena*, which already recognized as bio-fertilizers for the improvement of low land paddy soil fertility in Bangladesh and India [21] [22]. Therefore, *Azolla* spp. with Cyanobacterial inocula may be introduced in wetland paddy field as dual cropping with rice. *Azolla* spp. being hyper-accumulating heavy metals, may effectively remediate organic and inorganic wastes in contaminated land. In addition, *Sesbania* species may be introduced between rice-rice cropping pattern as green manuring leguminous crop after boro rice harvesting, which will improve soil fertility through influencing C Pool Index (CPI), and enhancing Carbon Management Index. The soil CMI, CPI and LI indexes reflect C sequestration and C cycling potential in different paddy ecosystems. In general, CMI value greater than one (>1) indicates that soil C is being rehabilitated, while CMI value smaller than one (<1) suggest that the system is degrading [23]. The higher CMI values in paddy soils are mainly contributed by the lability index (LI), which is closely related to the use of chemical fertilizers and exogenous organic amendments in the field. Labile organic carbon (LOC) is necessary for providing nutrient elements and energy for plant growth and microbial metabolism by its quick decomposition. It has been reported that soil CMI and SOC stocks with organic manure and crop residue N inputs treatments are higher than those with chemical fertilizer and no organic manure N inputs [24]. Currently, there is inadequate information on the suitable combinations of organic manures and inorganic fertilizers, biochars and biological amendments with *Azolla* cyanobacteria for improving degraded land, therefore, this research experiment was undertaken at two industrial locations of Mymensingh for restoration of contaminated land, reducing heavy metals pollution and mitigating cumulative GHGs emissions under suitable cropping patterns.

## 2. Materials and Methods

### 2.1. Experimental Sites

The experiment was conducted near BSCIC Industrial area, Maskanda, Mymensingh City Corporation, and Bhoradoba Union of Bhaluka Industrial area. The experimental soil was silty clay loam, with soil pH 6.1 - 6.3, SOM 2.1 - 2.3%. BRRI dhan-89 was cultivated during dry boro season (January 2022-May 2022, Jan 2023-May 2023).

### 2.2. Field Experimental Design, Layout and Treatments

The experiment was laid out in a randomized complete block design (RCBD) with three replications. The unit plot size was 20 m<sup>2</sup> (5 m × 4 m). Field experiments were carried out during January-May (Dry *Boro* season), 2022. In field conditions, the distance from row to row and hill to hill was 20 cm × 20 cm. Different combinations of soil amendments and chemical fertilizers are T1: NPKS (Farmers' practice) without any soil amendments, T2: N (50% RFD) with PKS + Tea waste bio-

char (5 t/ha), T3: N (50% RFD) with PKS + Vermicompost, VC (5 t/ha) with Silicate fertilizer (1.5 t/ha), T4: N (50% RFD) with PKS + Rice husk biochar (5.0 t/ha) with SF (1.5 t/ha), T5: N (50% RFD) with PKS + VC (5 t/ha) with Azolla Cyanobacteria dual cropping with rice, T6: N (50% RFD) with PKS + RHB (5.0 t/ha) with Azolla Cyanobacteria dual cropping with rice, T7: N (50% RFD) with PKS + Vermicompost (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T8: N(50% RFD) with PKS + RHB (5.0 t/ha) + Oyster shell biochar (1.5 t/ha) and T9: N (50% RFD) with recommended PKS + Tea waste biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha). Two cropping patterns such as *Boro* rice - fallow - *T.Aman* rice (**CP-1**): and *Boro* rice - *Sesbania rostrata* - *T.Aman* rice (**CP-2**) were followed at both locations.

### 2.3. Fertilizer and Organic Amendments Application

The land was fertilized with half of the recommended urea (212 kg·ha<sup>-1</sup>), triple super phosphate (TSP) 100 kg·ha<sup>-1</sup>, muriate of potash (MoP) 120 kg·ha<sup>-1</sup>, phospho-gypsum 2.5 t·ha<sup>-1</sup>, and zinc sulfate 4 kg·ha<sup>-1</sup>, respectively. The whole amount of TSP, MoP and silicate fertilizer were applied as a basal dose at the time of final field preparation. Urea was applied in three equal splits as top dressing at the tillering stage *i.e.* 21 days after transplanting (DAT), the maximum tillering stage (35 DAT), and before panicle initiation (55 DAT). Organic amendments such as vermicompost, tea waste biochar, rice husk biochar and Oyster shell powder were applied in field just one day before rice transplanting.

### 2.4. Determination of Soil Properties and Carbon Management Index (CMI)

After rice harvesting, soil organic carbon [25], total-N% (Micro-Kjeldahl method) [26], available P (Olsen method) [27], available S (Calcium chloride (0.15%) extraction method), exchangeable calcium (Ca), sodium (Na) and potassium (K) were extracted from soil using 1 M CH<sub>3</sub>COONH<sub>4</sub> solution and their concentrations were directly determined by flame photometer. CMI is used to represent the C dynamics of the soil and land system, which also indicates soil C rehabilitation status. Soil organic carbon (SOC) is a major determinant for the sustainability of agricultural systems. The changes in C pools (active or total) reflect the changes in an agricultural system. CMI was calculated using the formula:  $CMI = CPI \times LI \times 100$  [23]. Here,  $CPI = \frac{SOC \text{ in the treated soil sample}}{SOC \text{ in the reference soil sample}}$ ;  $LI = \frac{LOCs}{LOCr}$ ; where LOCs and LOCr are the labile C in soil sample and the labile C in reference soil, respectively. The lability of C (L) was calculated:  $L = \frac{KMnO_4 \text{ oxidizable C content}}{\text{Total organic C-KMnO}_4 \text{ oxidizable C content}}$ .

For pre-digestion, 5 g of soil was placed in a 500 ml beaker and 10 ml of HNO<sub>3</sub> was added for heavy metals analysis. The mixtures were left overnight for the initial oxidation of organic matter. Then the samples were heated on a hot plate at 120°C for 1 hour. After heating, 5 ml of H<sub>2</sub>O<sub>2</sub> was added to each sample, and heating was continued at 130°C for another 1 hour. For heavy metals analysis, after

cooling, these digested samples are filtered by using Whatman grade 42 filter paper. Thereafter, filtered paper was kept in separate places to avoid contamination. After filtering the mixer, pour it into a 50 ml plastic bottle and fill it with deionized water. The soil samples were vortexed 10/100 times using a vortex machine. For heavy metal identification from water, 50 ml of acidified water samples were filtered by using 125 mm filter paper (Whatman grade 42) and poured into a plastic bottle. After that, samples were again filtered by syringe filter (0.45  $\mu\text{m}$  HPPTFE) and then set into the ICP-MS sampling point by using a test tube. Filtered soil samples were ready for Inductively Coupled Plasma Mass Spectrometry (ICP-MS, SHIMADZU-2030LF) analysis.

## 2.5. Rice Yield Components and Grain Yield

Rice growth and yield characteristics etc. were recorded under different treatments and locations. Grain yield and straw yield per unit area were recorded after harvest.

## 2.6. Gas Sampling and Detection of CH<sub>4</sub> and N<sub>2</sub>O gases

Gas samples were collected from field plots once a week through Closed Chamber technique [15] [28]. A rectangular-shaped transparent acrylic glass chamber 40 cm  $\times$  60 cm  $\times$  110 cm was used to collect air samples. A saline set was fixed through a tiny hole in the upper portion of the chamber and made airtight with glue. The saline set had the key to open or close the path for air collection. The CH<sub>4</sub> emission from an irrigated rice field was calculated from the increase in CH<sub>4</sub> concentrations per unit surface area of the chamber for a specific time interval. Air samples were collected by using locked and open-systematized special syringes after the placement of the chamber over the source. Pre-evacuated glass vials of 20 ml were used to collect and preserve samples for analysis. The concentrations of CH<sub>4</sub> and N<sub>2</sub>O in the collected air samples were measured by Gas Chromatograph at Soil Science Lab, Bangladesh Rice Research Institute, Gazipur.

## 2.7. Estimation of Global Warming Potentials (GWPs)

To estimate the GWP, CO<sub>2</sub> is typically taken as the reference gas, and an increase or reduction in emissions of CH<sub>4</sub> or N<sub>2</sub>O is converted into “CO<sub>2</sub>-equivalents” by means of their GWPs. In this study, we used the IPCC factors to calculate the combined GWP for 100 years,  $\text{GWP} = 28 \times \text{CH}_4, \text{ kg CO}_2\text{-equivalents ha}^{-1}) + 265 \times \text{N}_2\text{O, kg CO}_2\text{-equivalents ha}^{-1}$ .

## 2.8. Soil Contamination Factor (CF), Soil Pollution Load Index (PLI) and Potential Ecological Risk Index (PERI)

Various indexes have been widely used to determine environmental risks of toxic elements in soils such as contamination factor (CF), enrichment factor (EF) and geo-accumulation index (Liu *et al.* 2014). Contamination factor is used to ascertain the level of soil contamination by heavy metals. CF is the ratio of the concen-

tration of each heavy metal to the baseline or background value (concentration in unpolluted soil).

$$CF = C_{\text{metal}}/C_{\text{background}}$$

Average shale value for each heavy metal was considered as background concentration value [29]. CF values for describing the contamination level are expressed as  $CF < 1$  = low contamination;  $1 < CF < 3$  = moderate contamination,  $3 \leq CF < 6$  = considerable contamination and  $CF > 6$  very high contamination [30] [31]. Pollution load index (PLI) is a multi-metal approach for an overall assessment of soil and sediment quality with respect to trace metals concentrations [32]. The pollution load index (PLI) for a single site is the  $n^{\text{th}}$  root of  $n$  number of multiplied together contamination factor (CF) values.

The PLI for a single site may be obtained as follows:

$$PLI = (CF_1 \times CF_2 \times CF_3 \dots \times CF_n)^{1/n};$$

where,  $n$  is the number of metals.

#### Potential Ecological Risk Index (PERI)

The potential ecological risk (RI) index proposed by Hakanson was used to evaluate the environmental impact of contaminants accumulated in bottom sediments [30]. The value RI was calculated using the following formulas:

$$\text{Potential Ecological Risk Index, } RI = \sum Er$$

$$\text{Ecological Risk Factor } Er = Tr \times CF$$

Here, TR indicates Toxic Response Factor, CF means Contamination factor.

## 2.9. Statistical Analysis

At first experimental data were entered into Microsoft Excel. Then analysis of variance (ANOVA) was performed through using R software (R-4.3.3, 2024 version). Duncan's multiple range test (DMRT) was conducted to identify statistically significant differences between group means at 5% significance level.

## 3. Results

### 3.1. Grain Yield

The experimental findings revealed that rice grain yield was significantly ( $p < 0.01$ ) influenced by organic amendments under two cropping patterns. Rice grain yield was recorded 3000 - 3200 kg/ha, and 3900 - 4100 kg/ha in chemical fertilized plots (T1) at BSCIC Industrial area and Bhaluka Industrial area, Mymensingh. Soil amendments with Vermicompost plus SF (T3), RHB + SF (T4), VC (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice (T5), RH biochar (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice (T6) significantly increased rice grain yield (10.0% - 13.8%) compared to chemical fertilized plots (T1) at both locations (Table 1). Incorporation of Sesbania as green manuring crop between rice-rice cropping system increased rice grain yield in all amended field plots compared to rice- fallow- rice cropping system.

**Table 1.** Rice grain yield, CH<sub>4</sub> and N<sub>2</sub>O fluxes, and total GWPs during boro rice cultivation under different combinations of organic inorganic and biological amendments (BSCIC Industrial area, Mymensingh City Corporation).

Treatment	Cropping pattern I: Boro rice-fallow-T. aman rice					Cropping pattern II: Boro rice-Sesbania rostrata-T. aman rice						
	Grain yield (kg-ha <sup>-1</sup> )	Cumulative CH <sub>4</sub> (kg CO <sub>2</sub> ha <sup>-1</sup> season <sup>-1</sup> )	GWPs for CH <sub>4</sub> (kg CO <sub>2</sub> ha <sup>-1</sup> )	Cumulative N <sub>2</sub> O (kg-ha <sup>-1</sup> season <sup>-1</sup> )	GWPs for N <sub>2</sub> O (kg CO <sub>2</sub> ha <sup>-1</sup> )	Total GWPs (kg CO <sub>2</sub> ha <sup>-1</sup> )	Grain yield (kg-ha <sup>-1</sup> )	Cumulative CH <sub>4</sub> (kg CO <sub>2</sub> ha <sup>-1</sup> season <sup>-1</sup> )	GWPs for CH <sub>4</sub> (kg CO <sub>2</sub> ha <sup>-1</sup> )	Cumulative N <sub>2</sub> O (kg-ha <sup>-1</sup> season <sup>-1</sup> )	GWPs for N <sub>2</sub> O (kg CO <sub>2</sub> ha <sup>-1</sup> )	Total GWPs (kg CO <sub>2</sub> ha <sup>-1</sup> )
T1: NPKS (Farmers' practice) without any amendments	3000	181	5070	0.490	136.2	5210	3200	193	5405	0.510	141.7	5546
T2: N (50%) with recommended PKS + Tea waste biochar (5.0 t/ha) + SF (1.5 t/ha)	3600	163	4560	0.350	106.0	4670	3700	170	4760	0.380	105.0	4870
T3: N (50%) with recommended PKS+ Vermicompost (5.0 t/ha) +SF (1.5 t/ha)	4100	160	4480	0.370	103.0	4585	4200	167	4676	0.410	103.0	4790
T4: N (50%) with recommended PKS + RH biochar (5.0 t/ha) + SF (1.5 t/ha)	3900	168	4710	0.310	101.0	4810	4000	173	4845	0.350	106.0	4950
T5: N (50%) with recommended PKS + VC (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice	4200	165	4620	0.420	117.0	4740	4400	168	4700	0.450	125.0	4825
T6: N (50%) with recommended PKS + RH biochar (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice	4000	170	4760	0.350	93.0	4855	4300	175	4900	0.400	106.0	5010
T7: N (50%) with recommended PKS + VC (5.0 t/ha) + Oyster shell biochar (1.5 t/ha)	3700	172	4816	0.390	106.0	4922	4000	183	5124	0.420	117.5	5240
T8: N (50%) with recommended PKS + RH biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha)	3800	175	4900	0.360	101.0	5001	3900	179	5012	0.390	103.5	5115
T9: N (50%) with recommended PKS + Tea waste biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha)	3300	169	4732	0.330	93.0	4825	3600	177	4960	0.360	108.0	5068
LSD	78.3	5.46	61.5	0.067	23.8	55.6	87.3	9.5	123.7	0.075	27.3	53.8
CV (%)	8.24	3.65	3.92	1.08	2.16	9.5	10.3	7.7	11.3	9.6	10.6	7.8
Level of Significance	*	**	**	**	***	**	*	**	**	**	**	**

Note: VC: Vermicompost, RHB: Rice Husk Biochar, T1: NPKS (Farmers' practice) without any amendments, T2: N (50%) with PKS + Tea waste, T3: N (50%) RFD with PKS + Vermicompost (5 t/ha) with silicate fertilizer (1.5 t/ha), T4: N (50%) RFD with PKS + Rice husk biochar (5.0 t/ha) with SF (1.5 t/ha), T5: N (50%) RFD with PKS + VC (5 t/ha) with Azolla cyanobacteria dual cropping with rice, T6: N (50%) RFD with PKS + RHB (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice, T7: N (50%) RFD with PKS + Vermicompost (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T8: N (50%) RFD with PKS + RHB (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T9: N (50%) with recommended PKS + Tea waste biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha).

### 3.2. Cumulative Methane (CH<sub>4</sub>) Emission during Rice Cultivation

This study revealed that seasonal cumulative methane emission was significantly ( $p < 0.01$ ) influenced by organic amendments. At BSCIC Industrial area of Mymensingh, the maximum cumulative CH<sub>4</sub> emission 181 - 193 kg/ha was recorded in chemical fertilized (T1) plot followed by 172 - 183 kg/ha in T7 (VC + Oyster shell), 175 - 179 kg/ha in T8 (RHB + Oyster shell biochar), 170 - 175 kg/ha in T6 (RHB + Azolla Cyano bacteria), 169 - 177 kg/ha in T9 (Tea waste biochar+ Oyster shell biochar), 168 - 173 kg/ha in (T4), 165 - 168 kg/ha in T5 (VC + Azolla Cyanobacteria), 163 - 170 kg/ha in T2 (Tea waste biochar + SF) and the least amount of seasonal CH<sub>4</sub> was found 160 - 167 kg/ha in T3 (VC + SF) treated field plots under Rice -Fallow-Rice (CP-1) and Rice-Sesbania-rice (CP-II) cropping patterns, respectively, (**Table 1**). On an average, the maximum decrease in cumulative CH<sub>4</sub> emissions were calculated 13.0%, 11.0%, 9.0%, 8.0% and 7.0% for T3 (VC + SF), T5 (VC + Azolla Cyanobacteria), T4 (RHB + SF), T6 (RHB + Azolla Cyanobacteria.) and T9 (Tea waste biochar + Oyster shell biochar), compared to chemical fertilizer treated plot (T1) under CP-I and CP-II.

At Bhaluka Industrial area, the maximum cumulative seasonal CH<sub>4</sub> emissions were recorded 193.0 - 198.0 kg/ha in chemical fertilizer treated (T1) plot. The decrease in cumulative CH<sub>4</sub> emissions were estimated 6.6% - 13.0% with soil amendments compared to chemical fertilizer treated plot (T1) under CP-I and CP-II. The maximum decrease in cumulative CH<sub>4</sub> emissions were observed 11.0% - 13.0% with Vermicompost and Azolla Cyanobacteria (T5), Tea waste biochar amendments with silicate fertilization (T2) and Vermicompost amendments with silicate fertilization (T3: VC + SF), respectively, compared to farmers practice (T1). The least seasonal cumulative CH<sub>4</sub> flux 168 - 173 kg/ha was recorded with VC amendments and silicate fertilizer application (T3, **Table 2**). Comparatively, higher seasonal cumulative CH<sub>4</sub> emission was recorded in Rice-Sesbania-rice Cropping system (CP-II) than that of rice -fallow-rice Cropping system (CP-1).

### 3.3. Cumulative Nitrous Oxide (N<sub>2</sub>O) Emission during Rice Cultivation

This experiment showed that cumulative nitrous oxide (N<sub>2</sub>O) emission was significantly influenced by the combinations of tea waste biochar, Vermicompost, silicate fertilizer, rice husk biochar amendments and inorganic nitrogen fertilizer application at two locations. In Mymensingh BSCIC area, the maximum cumulative seasonal N<sub>2</sub>O flux was recorded 0.460 - 0.510 kg/ha at farmers' practiced chemical fertilized plot(T1), which was decreased by 27.0% - 32.0% with rice husk biochar (RHB) amendments and Silicate fertilization (T4), 23.0% - 25.0% with Tea waste biochar amendments and silicate fertilizer application (T2) and 23% - 24.0% with Tea waste biochar and Oyster shell biochar (T9 under CP-I and CP-II (**Table 1**).

In case of Bhaluka experimental site, the maximum reduction in cumulative

seasonal N<sub>2</sub>O fluxes were recorded 33% - 36.0%, 31% - 35.0% and 29.0% - 32.0% with rice husk biochar (RHB) amendments and Silicate fertilization (T4), Tea waste biochar with silicate fertilization (T2), and rice husk biochar with Oyster shell biochar amendments (T8), respectively, under CP-I and CP-II (**Table 2**). On and average, the cumulative nitrous oxide (N<sub>2</sub>O) emissions were decreased by 27.0% - 36.0% with rice husk biochar (RHB) amendments and Silicate fertilization (T4), 23% - 35.0% with Tea waste biochar and silicate fertilization (T2), and 29.0% - 32.0% with rice husk biochar (RHB) and Oyster shell biochar amendments.

### 3.4. Total Global Warming Potential (GWP) for Rice Cultivation

Total global warming potential was significantly influenced by tea waste biochar, vermicompost, silicate fertilizer, Oyster shell biochar and rice husk biochar amendments in the contaminated land. At BSCIC experimental area of My-mensingh, the maximum total global warming potential (GWP) 5210 - 5546 kg CO<sub>2</sub> eq. ha<sup>-1</sup> was estimated in only chemically fertilized field plot (T1: Farmers practice), followed by 4950 - 5236 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T7: VC + Oyster shell biochar), 5000 - 5115 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T8: RH biochar + Oyster shell biochar), 4830 - 5070 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T9: Tea waste biochar + Oyster shell biochar), 4855 - 5010 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T6: RH biochar (5.0 t/ha) with Azolla cyanobacteria), 4810 - 4950 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T4: RH biochar with SF amendment), 4740 - 4830 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T5: VC (5.0 t/ha) with Azolla Compost cyanobacteria), 4670 - 4870 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T2: Tea waste biochar + SF), and the least GWP) was estimated 4585 - 4790 kg CO<sub>2</sub> eq. ha<sup>-1</sup> in VC amendments with SF (T3: Vermicompost + SF) (**Table 1**).

At Bhaluka Industrial site, the maximum GWPs 5450 - 5700 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T1), followed by 5155 - 5295 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T8), 5100 - 5250 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T7), 5030 - 5200 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T9), 5060 - 5156 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T5), 5060 - 5156 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T6), 5022 - 5140 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T4), 4980 - 5105 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T3), 4940 - 5080 kg CO<sub>2</sub> eq. ha<sup>-1</sup> (T5) and the least GWP 4800 - 5008 kg CO<sub>2</sub> eq. ha<sup>-1</sup> for Tea waste and SF amendment (T2) (**Table 2**). On average rice-sesbania-rice (CP-II) revealed higher GWPs values compared to rice-fallow-rice (CP-I) irrespective of soil amendments.

### 3.5. Soil Properties after Rice Harvest

Our results showed that soil amendments with Rice husk biochar, Tea waste biochar, Vermicompost with silicate fertilizer, Oyster shell biochar and Azolla cyanobacteria dual cropping with rice significantly increased soil pH, SOC contents, available P, available K and available N at both locations (**Table 3** and **Table 4**), indicating an improvement in soil fertility.

The increase in SOC in the amended field soils may be due to the high C/N ratio and cellulose contents in the added organic materials, compost and pyrolysed biochar. Carbon management index (CMI > 1) value more than one indicates improved carbon restoration status via Rice husk biochar (5.0 t/ha) with Azolla Cy-

anobacteria incorporation in field, VC amendments (5.0 t/ha) with Azolla Cyanobacteria incorporation, Tea waste biochar (5.0 t/ha) with Oyster shell powder and RHB (5.0 t/ha) with Oyster shell powder; whereas the least CMI value denotes the depletion of soil organic carbon materials (SOC), and raised turnover levels because of increased temperature and SOC depletion. The maximum improvement in overall soil properties was found with rice husk biochar and Azolla cyanobacteria incorporation, Vermicompost amendment with silicate fertilization and Azolla cyanobacteria incorporation in field soils, and Tea waste biochar amendments with silicate fertilizers alongwith half of the recommended N-fertilization.

### 3.6. Heavy Metals Contents in Post-Harvest Soil

The experimental findings revealed that heavy metals viz. Cu, Zn, Cd, Pb, and Mn contents in soil significantly varied, probably due to different agro-industrial sources in the selected locations (**Table 3** & **Table 4**). In addition, Tea waste biochar (5.0 t/ha) with Oyster shell powder and RHB (5.0 t/ha) with Oyster shell powder, Vermicompost and rice husk biochar (RHB) amendments alongwith Azolla Cyanobacteria dual cropping with rice influenced the heavy metals contents in post-harvest soil. At BSCIC Industrial area, Mymensingh, the heavy metals concentrations in farmers' field soils (T1: Farmers' practice, without any soil amendments) were found within the range 48.6 - 51.5 mg/kg, 67.3 - 69.5 mg/kg, 1.65 - 1.68 mg/kg, 47.3 - 48.7 mg/kg, and 1230 - 1380 mg/kg for Cu, Zn, Cd, Pb and Mn, respectively, (**Table 3**). The heavy metals contents in farmers' conventional practiced field plots at Bhaluka Industrial site were recorded 53.7 - 55.6 mg/kg, 54.7 - 58.5 mg/kg, 1.56 - 1.63 mg/kg, 49.3 - 52.7 mg/kg and 1380 - 1450 mg/kg for Cu, Zn, Cd, Pb and Mn, respectively (**Table 4**). In general, lower heavy metals contents were found in Boro rice-Sesbania-T. Aman cropping pattern compared to Boro rice-fallow-T. Aman rice cropping pattern. In addition, rice husk biochar with Azolla-Cyanobacteria and Oyster shell, Tea waste biochar with Oyster shell biochar amendments in selected field plots effectively decreased the contents of Cu, Zn, Cd, Pb, and Mn at both locations, although significant variations in the heavy metals concentrations were observed among the amended field soils at both locations. In the selected locations, Mn contents were found very high (T1: 1230 - 1450 mg/kg), which was significantly decreased by RHB with Oyster shell amendments (T8) and Tea waste biochar with Oyster shell amendments (T9). Lead (Pb) and Cadmium (Cd) contents in soil were also decreased significantly with rice husk biochar, tea waste biochar and vermicompost amendments with Oyster shell amendments (T7, T8 and T9) (**Table 3** & **Table 4**). In this experiment, the concentrations of Cd, Pb, and Mn were found significantly high compared to the World average concentration of Shale or background values [29], which may be due to high contamination factors for Cd, Pb and Mn.

In addition, improvement in soil redox status (Eh) and CMI were observed in the amended field soils at both locations (**Table 3** and **Table 4**) after rice harvest.

**Table 2.** Rice grain yield, CH<sub>4</sub> and N<sub>2</sub>O fluxes, and total GWPs during boro season rice cultivation under different combinations of organic and inorganic amendments (Bhaluka Industrial area, Mymensingh).

Treatment	Cropping pattern I: Boro rice-fallow-T. aman rice					Cropping pattern II: Boro rice-Sebania rostrata-T. aman rice				
	Grain yield (kg-ha <sup>-1</sup> )	Cumulative CH <sub>4</sub> (kg-ha <sup>-1</sup> ·season <sup>-1</sup> )	Cumulative N <sub>2</sub> O (kg-ha <sup>-1</sup> ·season <sup>-1</sup> )	GWPs for N <sub>2</sub> O (kg CO <sub>2</sub> -ha <sup>-1</sup> )	Total GWPs (kg CO <sub>2</sub> -ha <sup>-1</sup> )	Grain yield (kg-ha <sup>-1</sup> )	Cumulative CH <sub>4</sub> (kg-ha <sup>-1</sup> ·season <sup>-1</sup> )	Cumulative N <sub>2</sub> O (kg-ha <sup>-1</sup> ·season <sup>-1</sup> )	GWPs for CH <sub>4</sub> (kg CO <sub>2</sub> -ha <sup>-1</sup> )	Total GWPs (kg CO <sub>2</sub> -ha <sup>-1</sup> )
T1: NPKS (Farmers' practice) without any amendments	3900	189.0	0.550	153	5290	5290	153	5545	198.0	5705
T2: N (50%) with recommended PKS + Tea waste biochar (5.0 t/ha) + SF (1.5 t/ha)	4100	170.0	0.360	100.0	4760	4800	100.0	4900	175.0	5008
T3: N (50%) with recommended PKS + Vermicompost (5.0 t/ha) + SF (1.5 t/ha)	4600	173.0	0.410	138.0	4850	4980	138.0	4985	178.0	5105
T4: N (50%) with recommended PKS + RH biochar (5.0 t/ha) + SF (1.5 t/ha)	4400	175.0	0.350	122.0	4900	5022	122.0	5012	179.0	5140
T5: N (50%) with recommended PKS + VC (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice	4700	172.0	0.430	114.0	4820	4930	114.0	4960	177.0	5080
T6: N (50%) with recommended PKS + RH biochar (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice	4500	177.0	0.390	108.0	4956	5060	108.0	5050	180.0	5156
T7: N (50%) with recommended PKS + VC (5.0 t/ha) + Oyster shell biochar (1.5 t/ha)	4400	178.0	0.450	119.0	4985	5105	119.0	5125	183.0	5250
T8: N (50%) with recommended PKS + RH biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha)	4300	180.0	0.370	105.0	5050	5155	105.0	5180	185.0	5295
T9: N (50%) with recommended PKS + Tea waste biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha)	4200	175.0	0.470	131.0	4900	5031	131.0	5070	181.0	5203
LSD	120.6	2.6	0.18	18.3	154.7	487	18.3	785	9.7	133.7
CV (%)	13.3	10.6	0.09	9.3	7.5	10.5	9.3	11.6	8.6	11.5
Level of Significance	**	***	**	**	**	***	**	**	**	**

Note: VC: Vermicompost RHB: Rice Husk Biochar T1: NPKS (Farmers' practice) without any amendments, T2: N (50%) with PKS + Tea waste, T3: N (50%) with PKS + Vermicompost (5 t/ha) with silicate fertilizer (1.5 t/ha), T4: N (50% RFD) with PKS + Rice husk biochar (5.0 t/ha) with SF (1.5 t/ha), T5: N (50% RFD) with PKS + VC (5 t/ha) with Azolla cyanobacteria dual cropping with rice, T6: N (50% RFD) with PKS + RHB (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice, T7: N (50% RFD) with PKS + Vermicompost (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T8: N (50% RFD) with PKS + RHB (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T9: N (50%) with recommended PKS + Tea waste biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha).

**Table 3.** Soil properties, CMI, Heavy metals contaminations and soil pollution load (SPLI) index after soil amendments in the selected Industrial Sites (BSCIC Industrial area, Mymensingh City Corporation).

Treatments	Cropping pattern I: Boro rice-fallow-T. aman rice										Cropping pattern II: Boro rice-Sesbania rostrata-T. aman rice																			
	Soil pH	SOC	CMI	Heavy metals concentration (mg/kg)					Contamination Factor (CF)					PLI	PERI	Soil pH	SOC	CMI	Heavy metals concentration (mg/kg)					Contamination Factor (CF)					PLI	PERI
				Cu	Zn	Cd	Pb	Mn	Cu	Zn	Cd	Pb	Mn						Cu	Zn	Cd	Pb	Mn	Cu	Zn	Cd	Pb	Mn		
T1	6.5	14.7	0.91	51.6	69.5	1.68	48.3	1380	1.15	0.73	5.60	2.42	1.62	<b>1.79</b>	<b>188</b>	6.6	15.3	0.93	48.6	67.3	1.65	47.3	1230	1.08	0.71	5.50	2.37	1.45	<b>1.70</b>	<b>184</b>
T2	6.6	15.3	0.94	49.5	66.8	1.65	47.8	1320	1.10	0.70	5.50	2.39	1.55	<b>1.74</b>	<b>184</b>	6.6	16.6	0.97	47.3	63.6	1.61	44.6	1150	1.05	0.67	5.37	2.23	1.35	<b>1.63</b>	<b>179</b>
T3	6.7	16.6	0.98	46.3	63.5	1.67	44.6	1290	1.03	0.67	5.57	2.23	1.52	<b>1.67</b>	<b>185</b>	6.8	17.5	1.05	43.6	60.3	1.58	43.3	1100	0.97	0.63	5.27	2.17	1.29	<b>1.55</b>	<b>175</b>
T4	6.8	17.5	1.07	43.5	60.6	1.63	41.7	1250	0.97	0.64	5.43	2.09	1.47	<b>1.59</b>	<b>180</b>	6.9	18.6	1.15	41.5	58.7	1.55	39.7	1080	0.92	0.62	5.17	1.99	1.27	<b>1.49</b>	<b>171</b>
T5	6.7	18.6	1.15	45.6	58.3	1.59	38.6	1200	1.01	0.61	5.30	1.93	1.41	<b>1.55</b>	<b>175</b>	6.9	19.3	1.24	42.7	55.3	1.51	36.6	1020	0.95	0.58	5.03	1.83	1.20	<b>1.44</b>	<b>166</b>
T6	6.7	19.3	1.17	44.5	56.7	1.61	39.5	1130	0.99	0.60	5.37	1.98	1.33	<b>1.53</b>	<b>177</b>	6.9	20.6	1.28	41.3	52.6	1.48	37.6	950	0.92	0.55	4.93	1.88	1.12	<b>1.39</b>	<b>163</b>
T7	6.7	19.7	1.16	47.3	57.6	1.63	36.5	1100	1.05	0.61	5.43	1.83	1.29	<b>1.52</b>	<b>179</b>	7.0	21.3	1.26	46.6	54.3	1.50	34.5	930	1.04	0.57	5.00	1.73	1.09	<b>1.41</b>	<b>165</b>
T8	6.8	19.8	1.15	46.6	55.3	1.65	35.3	1050	1.04	0.58	5.50	1.77	1.24	<b>1.49</b>	<b>180</b>	6.9	19.6	1.23	45.3	53.5	1.52	33.5	900	1.01	0.56	5.07	1.68	1.06	<b>1.38</b>	<b>167</b>
T9	6.8	18.6	1.14	45.3	51.6	1.62	34.7	1030	1.01	0.54	5.40	1.74	1.21	<b>1.44</b>	<b>177</b>	6.9	19.7	1.21	44.6	50.3	1.49	31.7	860	0.99	0.53	4.97	1.59	1.01	<b>1.38</b>	<b>163</b>
LSD	0.19	3.3	0.24	4.7	6.1	0.07	6.3	17.5	0.21	0.18	0.35	0.23	0.15	0.43	9.5	0.11	3.7	0.22	4.8	13.6	0.13	6.3	21.6	0.19	0.13	0.28	0.45	0.15	0.48	11.3
CV (%)	6.5	4.7	6.3	7.8	8.3	4.3	8.5	9.3	6.3	4.5	8.6	7.8	6.5	7.3	10.3	4.6	6.7	6.5	5.7	6.5	5.6	9.5	12.3	9.3	7.8	6.5	7.5	8.6	6.7	9.7
Level of Significance	NS	*	*	*	*	NS	*	*	*	*	*	*	**	*	**	NS	*	*	*	*	*	*	*	*	*	*	*	*	*	**

Note T1: NPKS (Farmers' practice) without any amendments, T2: N (50% RFD) with PKS + Tea waste, T3: N (50% RFD) with PKS + Vermicompost (5 t/ha) with silicate fertilizer (1.5 t/ha), T4: N (50% RFD) with PKS + Rice husk biochar (5.0 t/ha) with SF (1.5 t/ha), T5: N (50% RFD) with PKS + VC (5 t/ha) with Azolla cyanobacteria dual cropping with rice, T6: N (50% RFD) with PKS + RHB (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice, T7: N (50% RFD) with PKS + Vermicompost (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T8: N (50% RFD) with PKS + RHB (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T9: N (50% RFD) with recommended PKS + Tea waste biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha).

**Table 4.** Soil properties, CMI, Heavy metals contaminations and soil pollution load (SPLI) index after soil amendments in the selected Industrial Sites (Bhaluka Industrial area, Mymensingh).

Treatments	Cropping pattern I: Boro rice-fallow-T.aman rice										Cropping pattern II: Boro rice-Sesbania rostrata-T.aman rice																			
	Soil pH	SOC (g.kg <sup>-1</sup> )	CMI	Heavy metals concentration (mg/kg)					Contamination Factor (CF)	SPLI	PERI	Soil pH	SOC (g.kg <sup>-1</sup> )	CMI	Heavy metals concentration (mg/kg)					Contamination Factor (CF)	SPLI	PERI								
				Cu	Zn	Cd	Pb	Mn							Cu	Zn	Cd	Pb	Mn				Cu	Zn	Cd	Pb	Mn			
T1	6.6	13.3	0.87	55.6	58.7	1.63	52.7	1450	1.24	0.62	5.43	2.64	1.71	1.80	185	6.7	15.3	0.97	53.7	54.7	1.56	49.3	1380	1.19	0.58	5.20	2.47	1.62	1.70	
T2	6.6	14.6	0.91	53.8	55.8	1.60	48.6	1330	1.20	0.59	5.33	2.43	1.56	1.70	180	6.8	16.3	1.05	50.3	50.3	1.53	45.6	1300	1.12	0.53	5.10	2.28	1.53	1.60	
T3	6.7	15.5	0.98	49.6	53.6	1.58	43.3	1260	1.10	0.56	5.27	2.17	1.48	1.60	176	6.8	16.5	1.10	48.6	48.6	1.51	40.3	1200	1.08	0.51	5.03	2.02	1.41	1.51	
T4	6.8	16.3	1.05	48.5	50.7	1.53	40.7	1180	1.08	0.53	5.10	2.04	1.39	1.53	170	6.8	17.6	1.15	45.3	45.7	1.48	36.5	1150	1.01	0.48	4.93	1.83	1.35	1.43	
T5	6.8	17.6	1.09	45.7	47.3	1.47	37.8	1120	1.02	0.50	4.90	1.89	1.32	1.44	163	6.9	18.3	1.19	41.5	43.6	1.43	33.6	1100	0.92	0.46	4.77	1.68	1.29	1.34	
T6	6.7	18.6	1.06	41.3	45.5	1.43	34.6	1080	0.92	0.48	4.77	1.73	1.27	1.36	158	6.9	19.6	1.20	38.3	39.5	1.40	30.3	1050	0.85	0.42	4.67	1.52	1.24	1.25	
T7	6.8	18.8	1.13	45.6	49.6	1.40	31.5	1050	0.97	0.52	4.67	1.58	1.24	1.37	154	6.9	20.6	1.24	40.6	38.6	1.35	29.7	1000	0.90	0.41	4.50	1.49	1.18	1.24	
T8	6.8	19.6	1.10	40.3	43.5	1.37	32.3	1100	0.90	0.46	4.57	1.62	1.29	1.31	151	6.9	20.3	1.20	37.5	37.3	1.34	30.5	1060	0.83	0.39	4.47	1.53	1.25	1.23	
T9	6.8	18.6	1.15	39.7	40.7	1.36	30.7	1050	0.88	0.43	4.53	1.54	1.24	1.27	150	6.9	19.6	1.25	38.3	39.6	1.33	28.3	1000	0.85	0.42	4.43	1.42	1.18	1.21	
LSD	0.11	3.7	0.22	4.8	13.6	0.13	6.3	21.6	0.19	0.13	0.28	0.45	0.15	0.48	15.3	0.11	3.7	0.22	4.8	13.6	0.13	6.3	21.6	0.19	0.13	0.28	0.45	0.15	0.48	
CY (%)	4.6	6.7	6.5	5.7	6.5	5.6	9.5	12.3	9.3	7.8	6.5	7.5	8.6	6.7	12.6	4.6	6.7	6.5	5.7	6.5	5.6	9.5	12.3	9.3	7.8	6.5	7.5	8.6	6.7	10.3
Level of Significance	NS	*	*	*	*	NS	*	*	*	*	*	*	**	*	**	NS	*	*	*	*	*	*	*	*	*	*	*	*	**	

Note T1: NPKS (Farmers' practice) without any amendments, T2: N (50%) with PKS + Tea waste, T3: N (50% RFD) with PKS + Vermicompost (5 t/ha) with silicate fertilizer (1.5 t/ha), T4: N (50% RFD) with PKS + Rice husk biochar (5.0 t/ha) with SF (1.5 t/ha), T5: N (50% RFD) with PKS + VC (5 t/ha) with Azolla cyanobacteria dual cropping with rice, T6: N (50% RFD) with PKS + RHB (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice, T7: N (50% RFD) with PKS + Vermicompost (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T8: N (50% RFD) with PKS + RHB (5.0 t/ha) + Oyster shell biochar (1.5 t/ha), T9: N (50%) with recommended PKS + Tea waste biochar (5.0 t/ha) + Oyster shell biochar (1.5 t/ha).

### 3.7. Soil Contamination Factor (CF) and Soil Pollution Load Index (SPLI)

The contamination factor for each heavy metal indicates the contamination level or degree of soil contamination. In case of Mymensingh BSCIC Industrial area, soil contamination factors (CF) were found within 1.08 - 1.15, (indicates moderate contamination) for Cu, CF 5.50 - 5.60 (very high contamination) for Cd, CF 2.37 - 2.42 for Pb (high contamination), and the CF for Mn was found 1.45 - 1.62 (high contamination) with farmers practiced filed plots (**Table 3**). Soil contamination factors for Cu, Pb, Cd and Mn were also found significantly higher in farmers' conventional practiced plot (T1) compared to the organic amended plots at Bhaluka Industrial site (**Table 4**).

Soil pollution load index (SPL) was estimated 1.44 - 1.79 and 1.27 - 1.80 at Mymensingh BSCIC and Bhakra Industrial areas (**Table 3 & Table 4**); the maximum value of PLI 1.70 - 1.80 (T1) indicates high soil degradation. Among the amendments, RHB with Oyster shell (T8), Tea waste biochar with Oyster shell (T9), Vermicompost with Oyster shell (T7) and RHB with Azolla Cyanobacteria dual cropping with rice (T6) showed lower pollution load index value than that of farmers' practice (**Table 3 & Table 4**). The decrease in pollution load with Rice husk biochar, Tea waste biochar, Vermicompost, and Oyster shell amendments may be due to the dissolution of metal pollutants.

### 3.8. Potential Ecological Risk Index (PERI)

At BSCIC Industrial area of Mymensingh the maximum potential ecological risk index 184 - 188 was obtained from farmers' practice without any soil amendments (**Table 3**), which reflects the high-risk category. Vermicompost with Azolla cyanobacteria, RHB with Azolla and Tea waste with Oyster shell amendments decreased the ERI (**Table 3**). In case of Bhaluka Industrial site maximum potential ERI value 176 - 185 was estimated from farmers practice (**Table 4**), which belongs to high-risk category. The potential ERI value was improved following Boro rice-Sesbania-T. Aman rice cropping pattern. Among the amendments, rice husk biochar, vermicompost and tea waste biochar with Oyster shell amendments significantly improved potential Ecological risk.

## 4. Discussion

Heavy metals pollution around Mymensingh including Bhaluka industrial areas has raised environmental and health concern due to industrialization and possible impact on agricultural crops, fisheries and humans. In this study, soil amendments with Tea waste biochar and silicate fertilization, Vermicompost and rice husk biochar with Azolla Cyanobacteria dual cropping in paddy field significantly decreased total cumulative CH<sub>4</sub> emission, N<sub>2</sub>O emission, and total GWPs under CP-I (Rice-fallow-rice) and CP-II (Rice-Sesbania-rice) during rice cultivation. Silicate fertilizer being potential source of electron acceptors, acted as an oxidizing agent, improved rice rhizospheric redox status (Soil Eh), thereby suppressed cu-

mulative CH<sub>4</sub> emissions. Azolla Cyanobacteria dual cropping in rice field reduced cumulative CH<sub>4</sub> and N<sub>2</sub>O emissions at both locations. The moderating effect of azolla-rice dual cropping on CH<sub>4</sub> emission could be due to photosynthetic release of oxygen by the floating azolla into the flooding water, which directly stimulated CH<sub>4</sub> oxidation at the soil-water interface and rhizosphere of the surface layer. In addition, azolla-rice dual cropping attributed to the large masses of floating plants, which acted as a physical barrier to the diffusion of CH<sub>4</sub> gas from anaerobic rice rhizosphere to the atmosphere. Vermicompost amendments in field and Azolla cyanobacteria dual cropping with rice significantly reduced seasonal CH<sub>4</sub>, whereas biochar amendments in paddy soil with Azolla Cyanobacteria dual cropping in rice field decreased N<sub>2</sub>O emissions, being acted as nitrification inhibitors. However, farmers practiced chemical fertilized (T1: Farmers' practice NPKS) field plots revealed higher CH<sub>4</sub> emission and N<sub>2</sub>O emission compared to other amended field plots. It has been reported that soil amendments with silicate fertilization with urea application decreased total seasonal CH<sub>4</sub> flux by 12% - 21% and increased rice grain yield by 5% - 18% in the upland rice paddy field [22]. Biochar amendments at 10 - 40 t·ha<sup>-1</sup> increased rice yields by 12% - 14% in unfertilized soils, and by 8.8% - 12.1% in soils with N fertilization, respectively [33]. In our field experiments, rice grain yield was increased by 10.0% - 13.8% with Vermicompost (5.0 t/ha) plus Silicate (T3), Rice husk biochar (5.0 t/ha) plus Silicate (T4), VC (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice (T5) and RH biochar (5.0 t/ha) with Azolla cyanobacteria dual cropping with rice (T6) compared to commonly used fertilized plots (T1). This may be due to the release of nutrients from the selected organic amendments, organic matter decomposition and improved soil properties, which enhanced nutrients uptake by rice plant, thereby contributed to yield increments.

The seasonal cumulative CH<sub>4</sub> emissions in this study were decreased by 9.0% - 16.5% with Vermicompost (5.0 t/ha) plus silicate fertilization (T3), Tea waste biochar (5.0 t/ha) plus silicate fertilization (T2), Vermicompost (5.0 t/ha) plus Azolla Cyanobacteria dual cropping with rice (T5), RH biochar with Azolla Cyanobacteria (T6) and RH biochar with silicate fertilizer (T4) amendments compared to chemical fertilized field plot (T1: 181 - 193 kg/ha) under CP-I and CP-II, respectively. The least amount of seasonal CH<sub>4</sub> emissions 160 - 163 kg/ha and 167 - 170 kg/ha were recorded with VC amendments plus silicate fertilization (T3), Tea waste biochar plus silicate (T2) and Vermicompost amendments Azolla cyanobacteria dual cropping with rice (T5) in BSCIC area of Mymensingh City Corporation. At Bhaluka Industrial area, the maximum decrease in cumulative CH<sub>4</sub> emissions were calculated 11.0% - 13.0% with Vermicompost (5.0 t/ha) plus silicate fertilization (1.5 t/ha), Tea waste biochar amendments (5.0 t/ha) with silicate fertilization (1.5 t/ha) and Vermicompost plus Azolla compost (5.0 t/ha) with Cyanobacterial inoculation in paddy field (T5), compared to farmers practice (T1: 193 - 198 kg/ha). The least seasonal cumulative CH<sub>4</sub> flux 168 - 175 kg/ha were recorded with Vermicompost plus silicate fertilization (T3) and Tea waste biochar

amendments with silicate fertilization (T2). This could be due to the electron accepting effects of silicate fertilizer, being acted as electron acceptors in the rice rhizosphere, thereby suppressed methanogenesis. It has been shown that Silicate fertilizer in combination with urea and *Azolla anabaena* decreased seasonal CH<sub>4</sub> flux by 12.0% and increased rice grain yield by 10.6%, while silicate fertilizer in combination with urea and cattle manure compost increased rice grain yield by 15.0% and decreased total seasonal CH<sub>4</sub> flux by 4.8% [34]. Furthermore, seasonal CH<sub>4</sub> emissions were decreased by 33.7%, 14.7% and 18.4% with slag-type silicate material amendments (2.0 t/ha) with recommended dose of NPK in rice fields of the Republic of Korea, Japan and Bangladesh, respectively [35]. It has also been reported that the rice plus *Azolla* without N fertilizer and with moderate N fertilizer (200 kg N ha<sup>-1</sup>) significantly reduced CH<sub>4</sub> emission by 12.3% and 25.3%, respectively, over the conventional fertilization (400 kg N ha<sup>-1</sup>) for rice cropping [36]. Dual cropping of azolla with rice significantly suppressed CH<sub>4</sub> emissions, probably due to an increase in dissolved oxygen concentration and redox potential at the soil-water interface [19]. It has also been shown that rice husk biochar application 5 - 10 t·ha<sup>-1</sup> significantly decreased cumulative CH<sub>4</sub> emission by 24% - 28.0% and N<sub>2</sub>O fluxes by 26% - 41.0% without a reduction in grain yield [37]. *Azolla anabaena* in combination with urea and silicate fertilization decreased total seasonal CH<sub>4</sub> flux by 12%, increased rice grain yield by 10.6% and improved overall soil properties [34].

In this study, the maximum reduction in cumulative N<sub>2</sub>O emission was recorded 29% - 32.0% with rice husk biochar (RHB) and silicate fertilization (1.5 t/ha), followed by 23% - 29.0% with tea waste biochar (5.0 t/ha) plus Oyster shell biochar (1.5 t/ha) and 24% - 27.0% with rice husk biochar (RHB) plus Oyster shell biochar (1.5 t/ha) and 23% - 25.0% with tea waste biochar (5.0 t/ha) and silicate fertilization (1.5 t/ha), respectively. The reduction in cumulative N<sub>2</sub>O emissions may be due to nitrification inhibition activities of rice husk biochar, tea waste biochar and Oyster shell biochar. It has been reported that nitrous oxide (N<sub>2</sub>O) emission was reduced by 28.3% - 33.8% from wetland paddy with basic slag application 1.0 - 2.0 t·ha<sup>-1</sup> in Indonesia [38]. Further increased application of basic slag (8 t·ha<sup>-1</sup>) reduced the N<sub>2</sub>O emissions by 20% - 66% from rice systems in China [39]. Biochar amendments in paddy soils of Japan and Bangladesh decreased seasonal cumulative N<sub>2</sub>O emissions by 31.8% and 20.0% respectively, followed by 26.3% and 25.0% reduction with biochar plus *Azolla*-cyanobacteria amendments [35]. The decrease in N<sub>2</sub>O emissions may be due to accelerated N immobilization, modified C/N ratio in soil and enzyme activity shown that biochar amendments at 40 t·ha<sup>-1</sup> increased total soil CH<sub>4</sub>-C emissions by 34% - 41% compared to the treatments without biochar and with or without N fertilization, respectively [33]. However, total N<sub>2</sub>O emissions were sharply decreased by 40% - 51% and by 21% - 28%, respectively in biochar amended soils with or without N fertilization.

The maximum global warming potential, GWP 5210 - 5546 kg CO<sub>2</sub> eq. ha<sup>-1</sup> and 5450 - 5705 kg CO<sub>2</sub> eq. ha<sup>-1</sup> were estimated in chemical fertilized field plots (T1:

Farmers' practice) at BSCIC Industrial and Bhaluka Industrial areas, respectively. GWPs were decreased by 9.0% - 14.0% with Vermicompost and Silicate fertilization (T3: VC + SF), Tea waste biochar and silicate fertilization (T2: Tea waste biochar + SF), and Vermicompost amendments with Azolla cyanobacteria dual cropping in rice paddy field (T5). The least GWPs 4585 - 4790 kg CO<sub>2</sub> eq. ha<sup>-1</sup>, and 4800 - 5008 kg CO<sub>2</sub> eq. ha<sup>-1</sup> were observed with Vermicompost and Silicate fertilization (T3), Tea waste biochar and silicate fertilization (T2) at BSCIC Industrial area and Bhaluka Industrial area, respectively. Calcium carbide, calcium silicate, and phosphogypsum amendments in paddy soil decreased total GWPs of CH<sub>4</sub> and N<sub>2</sub>O gases by 6.0% - 34.0%, whereas biochar amendments increased total GWP by 4.0% [40]. It has been shown that seasonal cumulative CH<sub>4</sub> emissions were significantly increased by 9.5% - 14.0% with biochar amendments, although global warming potentials were decreased by 8.0% - 12.0% with cyanobacterial inoculation plus biochar amendments [35]. The maximum decrease in GWP was calculated 22.0% - 30.0% with Azolla-cyanobacteria plus silicate slag amendments. The SOC level was significantly improved by organic amendments. Among the organic materials added/incorporated, Vermicompost, rice husk biochar, Azolla biomass, and Sesbania biomass input significantly increased the soil organic C (SOC). The increase of SOC content in the organic manure amended field soil was mainly due to the fact that the input of organic matter was higher than the amount removed. Therefore, higher SOC stocks in the amended field soil may be attributed due to higher organic materials inputs (RHB, Vermicompost, Azolla compost, Tea waste biochar, Oyster shell biochar) and lower rate of SOM breakdown and minimum losses of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O gases, compared to chemical fertilized field soils (T1: Farmers' practice), which eventually improved carbon management index (CMI > 1.0) in the amended field soils.

A significant variation of trace metals concentrations was observed under different soil amendments in the selected locations (BSCIC, Maskanda, Mymensingh and Bhoradoba, Bhaluka Industrial site). The concentration range for Cu 48.6 - 55.6 ppm, Zn 58.7 - 69.6 ppm, Cd 1.63 - 1.68 ppm, Pb 45.6 - 52.7 ppm, and Mn 1150 - 1350 ppm were recorded in the contaminated soils (T1) at the selected Industrial sites, which were found within the maximum permissible limit [41]. The results also showed that contamination factors for Cd (CF 5.43 - 5.60), Pb (CF 2.28 - 2.64) and Mn (CF 1.45 - 1.71) were significantly greater than for Cu and Zn, indicating that Cd, Pb and Mn were the main pollutants in the study sites. The high Mn concentrations (1250 - 1450 ppm) in contaminated soils (T1: Farmers' practice) may be due to high contamination factor, Industrial disposal, landfills, soil leaching and underground injection. In this study, the average concentrations of Cu and Zn were found 52.0 and 64.0 ppm in the contaminated soil (T1) at the selected locations, which were within the maximum permissible limit of 100 ppm [41]. Rice husk biochar, Tea waste biochar and Vermicompost amendments with Oyster shell powder effectively decreased the concentrations of Cd, Pb, and Mn in contaminated soils of the selected locations, probably due to the formation of

additional active sorption site, enhancing adsorption of potentially toxic metals [11]. Heavy metals concentrations in industrial affected soil samples at Sreepur Upazila of Gazipur district were found higher than the corresponding non-affected sites. The mean concentration of Cd, Pb, Cr, Ni, Cu and Zn in industrial affected soils were found 12.05, 101.10, 51.32, 20.79 and 55.40 mg·kg<sup>-1</sup>, respectively in the pre-monsoon, trace, 28.54, 40.96, 22.70, 2.46 and 7.72 mg·kg<sup>-1</sup>, respectively in the monsoon and 0.27, 10.49, 39.45, 20.69, 1.85 and 4.11 mg·kg<sup>-1</sup>, respectively in the dry season (42).

It has been reported that total Cd, Pb, Cu, Mn, and Zn contents in industrial areas of major cities of Bangladesh within the range 0.1 - 1.8, 17 - 89.7, 28 - 217, 106 - 177, and 53 - 477 mg·kg<sup>-1</sup>, respectively [43]. Cyanobacterial species (*Anabaena variabilis*) and microalgae (*Chlorella vulgaris*) significantly reduced heavy metals (Ca<sup>2+</sup>, Mg<sup>2+</sup>, Zn, Cr, and Mn) concentration from contaminated river water, due to removal of heavy metals through biosorption [44]. It has also been shown that VC (5.0 t/ha) and rice husk biochar (5.0 t/ha) amendments with silicate fertilization (1.5 t/ha) decreased the contents of Cu, Zn, Cd, Pb and Mn in contaminated soils, being supported by reduced toxicity of heavy metals to plants by soil-available silicon (Si) [45]. It was reported that Shell powder possess high remediation efficiency and adsorption capacity for the heavy metals in contaminated soils [46]. Rice husk biochar application reduced Cd and Pb contents in soil by 22.8% - 38.5% compared to the control at polluted sites [47]. Rice straw derived biochar amendments in paddy field showed the potentiality in enhancing SOC stocks over the two years' time period and the increased organic matter promoted the formation of organic bound Cd and improved the stability of Cd in soil [12]. It has been shown that biochar amendments decreased the exchangeable Cd content in soil and reduced (32%) the concentration factor for Cd uptake by rice [48].

In this study, introduction of *Sesbania* Spp between boro rice and aman season rice cultivation revealed higher rice grain yield, increased SOC contents, improved CMI value, lowered PLI and potential ERI value, which reflects better land quality and productivity compared to rice -fallow-rice cropping pattern, eventhough higher GWPs values were estimated in rice -sesbania -rice cropping pattern. It has also been reported that *Sesbania* spp accumulated heavy metals (Fe, Zn, Cu, Cr, Pb, Cd, Hg) from contaminated soils with industrial wastes in New Delhi, India [49].

The utilization of agro-industrial wastes for heavy metals remediation in contaminated sites may be feasible after considering the overall benefits of heavy metals removal capacity and resource utilization efficiency for environmental development. Therefore, prior to utilization of agro-industrial residues for heavy metals immobilization in contaminated field, it is necessary to evaluate the possible adverse environmental effects on each application site. In this study, the maximum soil pollution load index (SPLI) value was estimated 1.70 - 1.79 (BSCIC Industrial area, Mymensingh), 1.70 - 1.80 (Bhaluka Industrial area) for contaminated sites without any soil amendments, which indicates high soil degradation. The decrease in pollution loads through Rice husk biochar, Tea waste biochar and

Vermicompost amendments with Oyster shell may be due to the dissolution of metal pollutants.

The maximum potential ecological risk index 184 - 188 was obtained from farmers' practice (NPKS 100%, without any soil amendments, BSCIC area, My-mensingh), while ERI value 176 - 185 was estimated for Bhaluka Industrial site without any soil amendments, which indicates considerable ecological risk level. The potential ERI value was improved following Boro rice-Sesbania-T. Aman rice cropping pattern. Among the amendments, rice husk biochar, tea waste biochar and vermicompost with Oyster shell amendments significantly improved potential Ecological risk level (ERI).

## 5. Conclusion

The concentrations of Cd, Pb and Mn were found significantly high in the contaminated soils, which may be due to accumulation of industrial waste pollutants, hazardous landfills and contaminations factors above the acceptable level. Rice husk biochar, Tea waste biochar and Vermicompost amendments with Oyster shell significantly improved heavy metal contamination in selected locations. The maximum soil pollution load index (SPLI) value 1.70 - 1.80 for contaminated sites without any soil amendments, which indicates high soil degradation. In addition, the maximum potential ecological risk index 184 - 188 was obtained from farmers' practice without any soil amendments, which reveals considerable ecological risk level. The decrease in pollution loads and ERI through Rice husk biochar, Tea waste biochar and Vermicompost amendments with Oyster shell may be due to the dissolution of metal pollutants. Rice grain yield was increased by 10.0% - 13.8% with Vermicompost and silicate fertilization, rice husk biochar with SF, Vermicompost (5.0 t/ha) and rice husk biochar (5.0 t/ha) with Azolla cyanobacterial dual cropping with rice compared to fertilized plots at both locations. The mean maximum GWP 5380 - 5575 kg CO<sub>2</sub> eq. ha<sup>-1</sup> estimated from chemically fertilized field plot only (without any soil amendments), was significantly decreased by 14.5% - 15.7%, 11.5% - 14.5% and 7.5% - 12.5% with Vermicompost and silicate fertilization, Tea waste biochar and silicate fertilization and Vermicompost amendments with Azolla Cyanobacteria dual cropping in wetland paddy field ecosystem. Conclusively, integrated application of Vermicompost, rice husk biochar, Azolla compost, silicate fertilizer, tea waste biochar and Oyster shell amendments with half of the recommended N-fertilizer may be a feasible strategy to restore soil fertility, reduce heavy metals contamination, sustain rice productivity and mitigating GHGs emissions around industrial sites of Bangladesh.

## Ethical Declaration

This study does not involve any human or animals.

## Data Availability Statement

All data, tables, and figures in this manuscript are original.

## Authors' Contributions

Muhammad Aslam Ali: Being Supervisor and Principal Investigator, responsible for overall Research activities monitoring and helped in in draft write up.

Sk Md Fazlay Rabbi: Conducted field experiments and collected experimental data.

Hafsa Jahan Hiya: Compilation and validation of experimental data.

A. B. M. Shafiul Alam: Involved in Statistical data analysis and Tables preparation.

Sanjit Chandra barman: Organic amendments collection from different Organizations, Gas samples analysis.

Tanver Hossain: Field soil samples collection and Lab. works, especially heavy metals analytical works.

Zidan Ali Fagun: Helped in Gas Chamber placement in paddy field and samples collection from field during rice cultivation.

Sanjida Binta Shafin Mishu: Contributed to water sample analysis.

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## Conflicts of Interest

The authors declare that they have no conflict of interest regarding the publication of this paper.

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