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Cadmium contamination in agricultural soils of Bangladesh and management by application of organic amendments: evaluation of field assessment and pot experiments

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Abstract In recent years, cadmium (Cd) contamination in agricultural soils and its subsequent transfer to crops is one of the high-priority environmental and public health issues of global concern, especially in densely populated developing countries like Bangladesh. However, no effective strategy has been introduced or implemented yet to manage Cd-contaminated soils in order to sustain agricultural production with no human health risks. In this study, agricultural soil samples were collected from 60 locations of 10 upazilas from Tangail district to assess the extent of soil Cd contamination. The Cd concentration ranged from 0.83 to 4.08 mg kg^{-1} with a mean of 2.17 mg kg^{-1} in topsoil (0–15 cm), and from 0.67 to 3.74 mg kg^{-1} with a mean of 2.10 mg kg^{-1} in subsoil (16–30 cm). The values of contamination factor (CF) indicated that all the sampling locations were found to be highly contaminated with Cd. Pot trials with the application of different doses of biochar and vermicompost in Cd-contaminated soil (0.8 mg kg^{-1} Cd) revealed that integrated application of biochar (5 t ha^{-1}) and vermicompost (5 t ha^{-1}) was the best treatment that significantly ($p < 0.05$) reduced plant Cd concentration (72%) and increased the biomass of experimental crop, Red amaranth (*Amaranthus cruentus*). This combined treatment also

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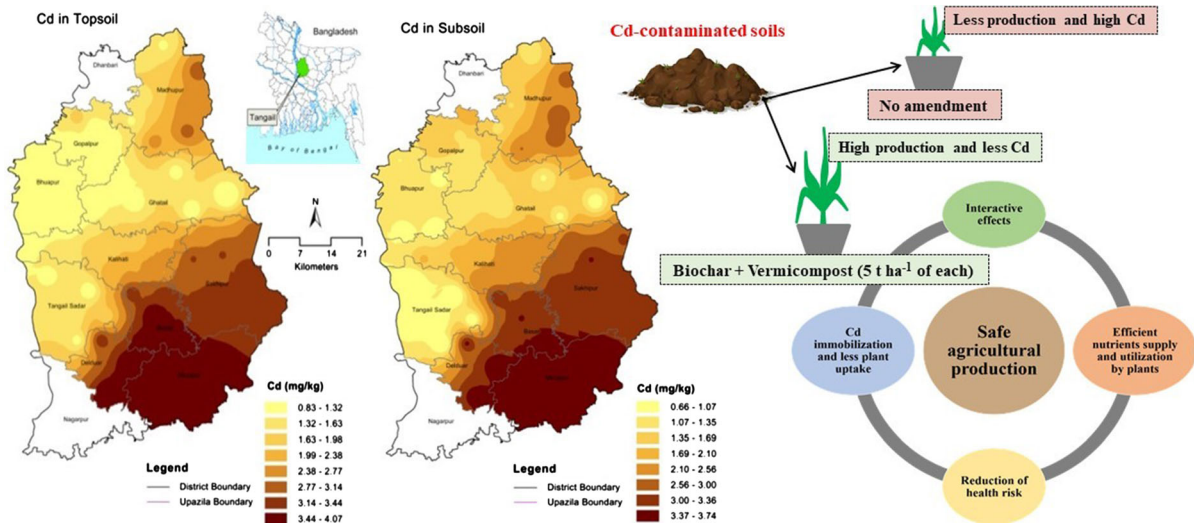
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significantly reduced the uptake of Cr (37%) when co-contamination was present. The study suggests the application of biochar (5 t ha^{-1}) in combination with vermicompost (5 t ha^{-1}) to reduce human health risk and increase crop production when the soil is loamy sand in texture.

Graphical abstract

considered to be the major source of Cd as well as other metallic contamination in agricultural soils (Atafar et al. [2010]; Meherg et al. [2013]). Among the inorganic fertilizers, phosphate fertilizers are used intensively for alleviating phosphorus (P) deficiency which has been reported in about 43% of the global soils (Liu et al. [2012]). Phosphate fertilizers are produced from non-renewable phosphate rocks (Cor-

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dell et al. [2009]), some of which also contain appreciable quantities of non-essential toxic heavy metals (Gambuś and Wieczorek [2012]; AlKhader [2015]; van der Wiel et al. [2016]). Taylor ([1997]) and Taylor et al. ([2007]) reported that the intensive use of phosphate fertilizers has even tripled the average concentration of Cd in agricultural soils (Background or unfertilized soil—0.16 mg Cd kg^{-1} soil, cropping soil—0.24 mg Cd kg^{-1} soil, pasture- 0.43 mg Cd kg^{-1} soil, horticulture—0.50 mg Cd kg^{-1} soil). Gambuś and Wieczorek ([2012]) found that 1 kg of P fertilizer contains 13 mg Cd, 60 mg Cr, 26 mg Cu, and 13 mg Pb.

The Cd, Cr and Pb often coexist in contaminated soils and are transferred from soil to plants (Shanker et al. [2005]; Jiang et al. [2012]). The Cd occurs naturally as a contaminant in all phosphate rocks. Rock phosphates originating from igneous rocks generally contain Cd concentrations of 0.7–30 mg kg^{-1} P (Oosterhuis et al. [2000]), whereas rock phosphate refined from sedimentary rocks may contain Cd concentrations higher than 556 mg kg^{-1} P (Mar and Okazaki [2012]). Accumulation of Cd along with other

Introduction

Globally, cadmium (Cd) contamination in soils has become a serious concern for environmental and human health, especially in developed countries experiencing rapid industrialization and practicing intensive agriculture (Zhao et al. [2010]; Cai et al. [2019]). Bangladesh, a heavily densely populated developing country, is well known for metals and metalloids pollution (Islam et al. [2018a]). Meharg et al. ([2013]) surveyed 12 countries from 4 different continents and found that the Cd concentration in rice of Bangladesh was the highest than that of USA, Japan, France, Italy, Spain, Cambodia, Ghana, India, Nepal, Sri Lanka, and Thailand. Among all the anthropogenic influences, repeated and excessive application of inorganic fertilizers along with pesticides is

toxic metals in agricultural soils leads to the transfer of these metals to the crops and may pose health risks to humans and animals (Mamun et al. 2016, 2017; Sultana et al. 2017). In addition, the recurring and excessive application of inorganic phosphate fertilizers may lead to accumulation of P in the surface soil since plants uptake only 10–15% of the P applied, termed as low P-use efficiency of plants, and the residual P is believed to be fixed in the soil as unavailable forms (Roberts and Johnston 2015), which subsequently causes eutrophication in water bodies through P runoff and leaching (Soltangheisi et al. 2018) and forces farmers to use more P fertilizers.

Researchers have found that organic amendments (e.g., biochar, compost, vermicompost, lignite and animal manure) are useful to improve soil quality and to stimulate the plant growth as well as to immobilize heavy metals in soils (Naeem et al. 2006; Herencia et al. 2007; Jouquet et al. 2011; Simmler et al. 2013). Organic amendments may increase soil pH and cation exchange capacity (CEC) and immobilize toxic metals in soils by binding metals to the adsorption sites and forming insoluble organometallic compounds that ultimately reduce the plant uptake of toxic metals (Pusz 2007; Hamid et al. 2018). Moreover, as Cd is a chalcophile element, it strongly binds with organic sulfur groups, R-SH, R-S-R, R-SS-R and heterocyclic S (Kabata-Pendias and Mukherjee 2007) and subsequently the concentration of Cd is decreased in soil solution. Several studies have been conducted to evaluate the effects of different organic amendments on crop production and management of metal-contaminated soils (Castaldi et al. 2005; Gadepalle et al. 2008; Angelova et al. 2010; Bian et al. 2014; Hamid et al. 2018). However, the use of organic amendments alone may not be enough to meet the enormous nutrient requirements of present-day high-yielding cultivars (Sanni and Ewulo 2015). On the other hand, very limited consensus is available on the benefits of combined application of organic amendments for maximizing the production of crops, increased immobilization of heavy metals, and soil nutrient management, especially in loamy sand soils. Adequate information is also not available regarding the optimum application rate of organic amendments for crop production in loamy sand soil contaminated with Cd and other metals.

Red amaranth (*Amaranthus cruentus*) is a widely consumed leafy vegetable in Bangladesh which serves as an alternative source of nutrition for people in developing countries like Bangladesh since it is highly nutritious and inexpensive source of carotenoid, protein, vitamins, and dietary fiber (Shukla et al. 2006; Gayathri and Anitha 2018). It has a growing period of 5–6 weeks thus making it an advantage for the rural and peri-urban farmers to keep cultivating it two or more times on the same piece of land in a year (Adewole and Igberaese 2011). Moreover, unlike other leafy vegetables, vegetable amaranth can be cultivated during hot summer months, even when no other green vegetables are available in the market, under varied soil and agro-climatic conditions (Gayathri and Anitha 2018), and about 80% of the harvested amaranth plants are edible (Oke 1980). In Bangladesh, its cultivation is increasing day by day and the total production is about 1500 thousand tons per year, while 70% of the plant is produced in winter season and 30% in summer season (BBS 2012). The potentiality of cultivating *A. cruentus* is high in Bangladesh because of its taste and flexibility of growing season and aforementioned price. In addition, leafy vegetable red amaranth thrives well on soils with high organic matter (Akinbile et al. 2016). Though a number of different studies reported optimum fertilizer amounts for the maximum production of *A. cruentus*, the results are sometimes substantially different as well as limited information is available on the synergistic and antagonistic effects of organic amendments with or without phosphate fertilizers on its production.

Thus, a field assessment was conducted to assess the level of Cd contamination in agricultural soils in a typical agricultural region of Bangladesh. Thereafter, experimental pot trials were performed to investigate the effects of biochar and vermicompost (as organic amendments), individually and in combination, with or without TSP (triple super phosphate) on the plant growth and yield, P content, and uptake of Cd and other metals (Cr and Pb) by red amaranth (*A. cruentus*) under different rate of application. Finally, a health risk assessment was done to evaluate the efficacy of the organic amendments for reducing the health risk of Cd and other metals (Cr and Pb) via dietary intake of produced *A. cruentus*.

Materials and methods

Study area and sampling sites

The study focused on the agricultural soils in Tangail district, which is located in the central part of Bangladesh (Fig. 1) and covers an area of 3414 km² consisting of 12 upazilas (Banglapedia 2020). Tangail district is characterized by four agro-ecological zones (AEZs), namely Active Brahmaputra-Jamuna Flood-plain (AEZ-7), Young Brahmaputra and Jamuna Floodplain (AEZ-8), Old Brahmaputra Floodplain (AEZ-9) and Madhupur Tract (AEZ-28) (Kumar et al. 2018). In the first three AEZ areas, mainly rice and vegetables are grown and most of these lands remain under water during April to August, sometimes even till September and Mid October. In Madhupur Tract, mainly pineapple and vegetables are grown. The areas have a tropical monsoon climate. The average minimum temperature is 11.4 °C observed in January, and the average maximum temperature is

recorded as 33.8 °C in April (BBS 2018), with an annual average rainfall of 1848 mm (BBS 2018) and annual average relative humidity of 80% (Akter et al. 2016). Rainfall starts mainly in April and continues until October, while about 65% of rainfall occurs during the monsoon season (June–September) (BBS 2018). According to the Department of Agricultural Extension (DAE), the district comprises 328,160 ha of agricultural land (MPSSWRD 2005) and produces about 847,309 tons of agricultural crops every year (Rahman and Mian 2016). Rice is the principal agricultural crop (65.7%), followed by oil seeds, wheat/maize, pulses, vegetables, spices, sugarcane, potato, mustard, and others (MPSSWRD 2005).

Soil sample collection and analysis

Although Tangail district is composed of 12 distinct upazilas, soil samples were collected from 10 upazilas for the current research which ensured that all the cropping patterns existing in Tangail district were
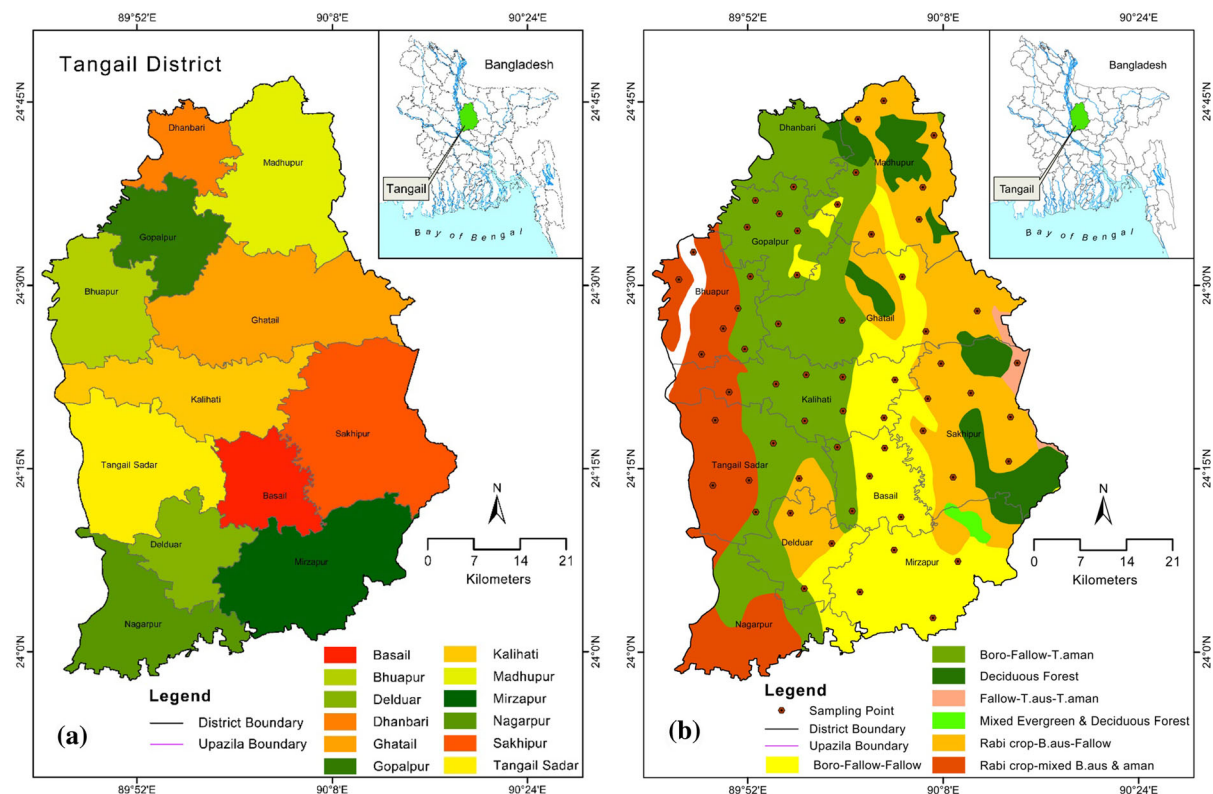
Fig. 1 Map showing the upazilas of Tangail district **a** and the sampling points at different upazilas in Tangail district of Bangladesh **b**. Colorful rectangles (in legend) are representing different upazilas **a** or cropping patterns **b** in the study area

sampled. Soil samples were collected from 60 different agricultural lands from the selected 10 upazilas of Tangail district, while the agricultural lands were selected considering various cropping patterns in the study area (Fig. 1). Both the topsoil (0–15 cm) and subsoil (16–30 cm) were collected from each sampling site. In each sampling site, three sampling points (about 5 feet distance) were selected. From each point, one top soil and one subsoil were collected. The three subsoils from three points were mixed together to form a representative composite subsoil sample. Similarly, composite top soil samples were prepared for each sampling site. In total, 120 samples (60 topsoil and 60 subsoil) were collected from the study area. Collected samples were dried at room temperature for 2 weeks then crumbled using a porcelain mortar, sieved through a 2 mm nylon mesh, and homogenized. The samples were finally stored in clean Ziploc bags under freezing conditions until chemical analysis.

The soil pH was determined using a digital pH meter (Hannah, Woonsocket, RI, USA) in a soil/water ratio of 1:2.5. Total P was determined colorimetrically using a spectrophotometer at 490 nm by developing yellow color with vanadomolybdate after collecting the extract by digestion with ternary acid mixture (conc. HNO_3: conc. HSO_4: $HClO_4$ (60–62%) = 5:1:2). To get the extract, 0.5 g of dried and ground sample was taken in a 50 ml conical flask and 10 ml ternary acid mixture was added. The mixture was allowed to predigest for 24 h, heated over a low gas flame, gradually raised the heat until the mixture becomes clear. Afterward, the mixture was allowed to cool and distilled water was added to make the volume up to 50 ml (Huq and Alam 2005).

For determining soil Cd concentrations, soil samples were wet-digested with aqua regia (HNO_3:-HCl = 1:3) mixture under closed system (Blum et al. 1996). For analysis, 1 g of soil sample was digested with 20 ml of aqua regia and heated until the acid is dried out. Then the mixture was diluted and volumed with distilled water. The digested samples were filtered with Whatman no. 42 filter paper. Finally, the extract was measured using AAS (Shimadzu AA-6800, Kyoto, Japan) at 228.8 nm (Huq and Alam 2005). Calibration curve was prepared using the certified standard solution of AAS (Wako Pure Chemical Industries, Ltd., Osaka, Japan). In Bangladesh, there is no prescribed permissible limits given by any governmental or non-governmental agencies for

Cd concentrations in soils. Thus, this study used the permissible limit of Cd in soil as 1.0 and 1.4 mg kg^{-1} according to the Dutch soil quality standard (MVROM 2000) and Canadian environmental quality standard (CCME 2003), respectively.

The level of contamination was calculated using the contamination factor (CF) by the following formula as used by several researchers (Islam et al. 2015a; Tusher et al. 2017):

$$CF = \text{Measured Cd concentration in soil/Background concentration of Cd} \tag{1}$$

The level of contamination can be classified based on the calculated CF values as: low (CF < 1), moderate ($1 \leq CF < 3$), considerable ($3 \leq CF < 6$), and high ($CF \geq 6$). The background value of Cd was considered as 0.11 mg kg^{-1} for Bangladesh soil (Kashem and Singh 1999).

Design of pot experiment

A pot experiment was carried out using black plastic buckets of 20 cm in diameter and 30 cm in height. Fifteen treatments of organic amendments and TSP, individually or in combination, including a control (soil without amendments) were used to evaluate their effects on plant growth and Cd uptake. The details of the treatments are described in Table 1. Biochar and vermicompost were used as organic amendments, while TSP was used as inorganic phosphate fertilizer. All the experiments were conducted in triplicate, and thus a total of 45 pots were used for the experiment. All the experimental pots were supplied with the same contaminated agricultural soil, vegetable seeds and irrigated with tap water. The basic properties of the biochar, vermicompost, and experimental soil used in the pot experiment are shown in Table S1. Since co-contamination of Cr and Pb was found in the experimental soil though not high enough (Table S1), the effects of studied treatments were also assessed on plant uptake of Cr and Pb along with Cd to evaluate how the treatments will influence the uptake of these toxic metals posing potential risks to human health. The pots were prepared with 7 kg of homogeneous soil mixed with different types of treatments. Finally, each of the experimental pots was labeled with treatment identification number and rate of amendment applied. The biochar, vermicompost and

Table 1 Details of treatments used in the pot experiment

Treatment ID	Name of treatment
Control	Control (Soil with no amendment)
T1	Biochar (1 t ha^{-1})
T2	Biochar (5 t ha^{-1})
T3	Biochar (10 t ha^{-1})
T4	Biochar (20 t ha^{-1})
T5	Vermicompost (1 t ha^{-1})
T6	Vermicompost (5 t ha^{-1})
T7	Vermicompost (10 t ha^{-1})
T8	Vermicompost (20 t ha^{-1})
T9	Biochar (0.5 t ha^{-1}) + Vermicompost (0.5 t ha^{-1})
T10	Biochar (2.5 t ha^{-1}) + Vermicompost (2.5 t ha^{-1})
T11	Biochar (5 t ha^{-1}) + Vermicompost (5 t ha^{-1})
T12	Biochar (1 t ha^{-1}) + TSP (40 kg ha^{-1})
T13	Biochar (5 t ha^{-1}) + TSP (60 kg ha^{-1})
T14	Biochar (10 t ha^{-1}) + TSP (80 kg ha^{-1})

TSP were collected from the local fertilizer shop of Tangail sadar upazila. The collected biochar was produced from sawdust at 350 °C temperature, and vermicompost was produced from cow dung by using earthworms.

Cultivation of experimental plant

Leafy vegetable *A. cruentus* was cultivated for the pot experiment. The *A. cruentus* was selected since it grows easily and is commonly consumed by people as an alternative source of nutrition, especially in developing countries, owing to low price and high nutritional value (Gayathri and Anitha 2018). Fresh and good quality seeds of *A. cruentus* were collected from the local seed shop of Tangail sadar upazila. The seeds were sown during winter in February 2019. Germination of seed started within 3 days of sowing. On the 5th day, maximum seeds were germinated. Intercultural operations such as watering and weeding were done to provide favorable conditions for the development of seedlings. Weeding was done seven days after sowing and repeated when necessary to keep the pots free from weeds. After one week of sowing, three plants with maximum growth were kept in each pot and the rest were removed.

Plant sample collection and analysis

The *A. cruentus* plants were harvested at maturity 55 days after sowing. At the time of harvesting, plants height from each pot were measured. Harvested plant samples were kept in Ziploc bags and properly labeled with the identification number. The plant samples were then washed with deionized water several times as like as the vegetables are washed before cooking to eliminate dust and dirt. The harvested plant samples were cut down into small pieces, and the fresh weight biomass of plants was measured (after drying with white paper towel) within 1–2 h of harvesting using a digital balance. The plant samples were then oven-dried at 75 °C for 48 h, and finally the dry weight biomass was measured. The dried vegetable samples were homogenized by grinding using a ceramic coated grinder. Ground samples were passed through a 2 mm Nylon sieve. The processed plant samples were analyzed for P content following the similar procedures used for soil samples, while the total Cd, Cr, and Pb concentrations were measured using AAS at 228.8, 357.9, and 283.3 nm, respectively, as also used by other researchers (e.g., Jamali et al. 2006, 2009; Sahito et al. 2016). Before analyzing the samples, extraction and digestion solution as well as method blanks in triplicate were analyzed as a part of standard quality control procedure and deducted the blank reading. Certified Reference Materials for soil (International Soil analytical Exchange—ISE 921) and plant

(International Plant analytical Exchange IPE 100) from Wageningen University, The Netherlands, were analyzed, and recoverable concentrations of the Certified Reference Materials were within 85 to 115% of the certified values. It is to be noted that the number of parameters determined for field assessments was not the same as the ones determined for the pot trials, while the results obtained by pot experiments might be different in field conditions. Thus, these can be considered as the shortcomings of this study.

Assessment of health risk of heavy metals

Calculation of metal concentrations in fresh weight

The concentrations of heavy metals in fresh vegetables (C_{FW}) were calculated by the following equation (Guo et al. 2019):

$$C_{FW} = C_{DW} \times (1 - w) \tag{2}$$

where C_{FW} is the concentration of heavy metals in fresh vegetables (mg kg^{-1}), C_{DW} is the concentration of heavy metals in dry vegetables (mg kg^{-1}), w is the water content in fresh vegetables which was measured by the following equation:

$$w = (\text{Fresh weight} - \text{Dry weight})/\text{Fresh weight} \tag{3}$$

Estimated daily intake of heavy metals

The estimated daily intake (EDI) of heavy metals via vegetable consumption was calculated by the following equation (Islam et al. 2015b):

$$EDI = (C_{FW} \times IR)/BW \tag{4}$$

where EDI is the estimated daily intake of heavy metals (mg day^{-1}), C_{FW} is the concentration of heavy metals in fresh vegetables (mg kg^{-1}), IR is the vegetable ingestion rate (kg day^{-1}), and BW is the body weight of individual consumer (kg). The IR was considered as 0.155 and 0.106 kg day^{-1} for adults and children, respectively, while the BW was considered as 60 and 16 kg for adults and children, respectively, as used in previous studies (Islam et al. 2015b; Shaheen et al. 2016).

Target hazard quotient (THQ)

The target hazard quotient (THQ) is an assessment of non-carcinogenic human health risk due to the exposure of heavy metals. The THQ for each heavy metal was calculated by the following equation (USEPA 1989):

$$THQ = \{(C_{FW} \times IR \times EFr \times ED)/ (RfD_o \times BW \times AT)\} \times 10^{-3} \tag{5}$$

where THQ is the target hazard quotient (dimensionless), EFr is the exposure frequency (365 days year^{-1}), ED is the exposure duration (70 years) equivalent to the average human lifetime, AT is the averaging time for non-carcinogens (365 days year^{-1} × ED), and RfD_o is the oral reference dose of individual metal (considered 0.0005, 1.5 and 0.0035 mg kg^{-1} day^{-1} for Cd, Cr and Pb, respectively, according to USEPA (2016)).The THQ ≥ 1 indicates that the exposed population will likely to experience long-term non-carcinogenic effects, while THQ < 1 indicates that the population is assumed to be safe (Wang et al. 2005).

Hazard index (HI)

Hazard index (HI) is the estimation of overall possible non-carcinogenic risk posed by more than one metal and was calculated by the following equation (USEPA 1989):

$$HI = \sum THQ = THQ\ metal\ 1 + THQ\ metal\ 2 + \dots + THQ\ metal\ n \tag{6}$$

where HI is the hazard index (dimensionless). When HI < 1, the vegetable is safe for human and is considered to be acceptable, while HI > 1 may pose non-carcinogenic risk and adverse health effects (USEPA 1989).

Target carcinogenic risk (TR)

Target carcinogenic risk (TR) is the assessment of lifetime cancer risk derived from exposure to heavy metals and was calculated by the following equation (USEPA 2006):

$$TR = \{(C_{FW} \times IR \times EFr \times ED \times CSF_o)/ (BW \times AT)\} \times 10^{-3} \tag{7}$$

where TR is the target cancer risk over a lifetime (dimensionless), CSF_o is the oral carcinogenic slope factor $(mg\ kg^{-1}\ day^{-1})^{-1}$, and AT is the averaging time for carcinogens (365 days $year^{-1} \times ED$). The CSF_o was considered 15 and 0.5 $(mg\ kg^{-1}\ day^{-1})^{-1}$ for Cd and Cr according to California Environmental Protection Agency (Zeng et al. 2015), while 8.5×10^{-3} $(mg\ kg^{-1}\ day^{-1})^{-1}$ for Pb (USEPA 2016). The Cd and Cr are classified as known human carcinogens (Group 1), and Pb is listed as probable carcinogen (Group 2A) by the International Agency for Research on Cancer (IARC 2011).

Statistical analysis

The collected data were subjected to descriptive statistics performed using MS Excel version 2013.

Spatial distribution of Cd in topsoil and subsoil was represented using ArcGIS 10.3.1. The analysis of variance (ANOVA) with least significance difference (LSD) post hoc test at 5% level of significance ($p < 0.05$) as well as test of homogeneity of variances was done using SPSS 17.0 to compare the effects of different treatments. Additionally, Pearson's correlation among organic amendment application, plant growth and P content as well as heavy metal concentrations was determined using SPSS 17.0. The results of Pearson's correlation were then depicted with a heat map using MS Excel 2013.

Results

Properties of agricultural soils and level of cadmium (Cd) contamination

The pH of subsoil was higher than that of top soil at all sampling sites (Fig. 2). The mean pH was 5.9 in the
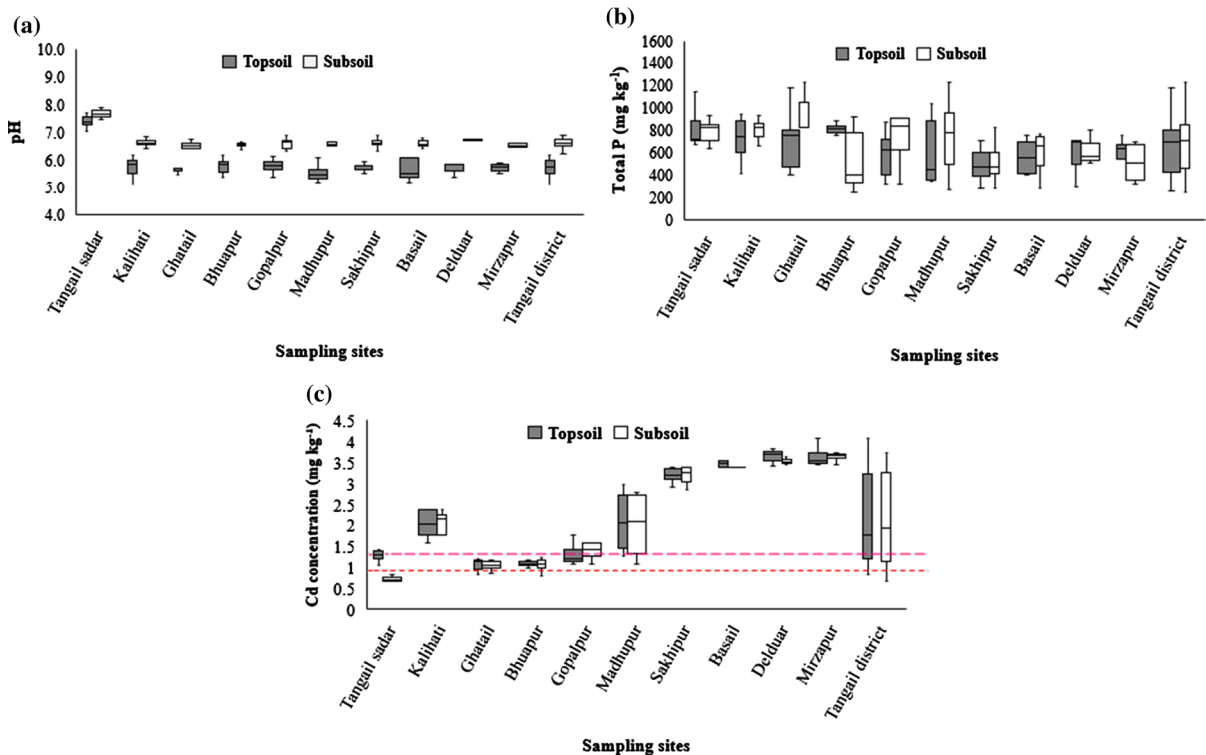
Fig. 2 Box-plots showing soil pH a, total P b, and Cd concentrations c in different upazilas along with Tangail district. All the values of different sampling sites were grouped to represent the overall scenario of Tangail district. The red and pink dotted lines in c are indicating the permissible limit of Cd in soil as 1.0 and 1.4 mg kg^{-1} according to the Dutch soil quality standard (MVROM 2000) and Canadian environmental quality standard (CCME 2003), respectively

topsoil and 6.7 in the subsoil in the Tangail district (Fig. 2a). In the study area, the topsoil pH ranged from 4.4 to 7.7 and from 6.2 to 7.9 in the subsoil. Similar levels of topsoil and subsoil pH were observed at different sampling sites excluding Tangail sadar upazila where higher pH values were observed in both topsoil and subsoil compared to other upazilas (Fig. 2a). On the other hand, the total P contents in topsoil and subsoil were observed to be variable and ranged from 261.39 to 1184.23 mg kg^{-1} (topsoil) and from 239.45 to 1481.07 mg kg^{-1} (subsoil) in the study area. Among the sampling sites, the mean total P contents were found to be higher in topsoil of the 4 sampling sites (Tangail sadar, Bhuapur, Sakhipur, and Mirzapur) than the mean total P contents in subsoil, while opposite results were observed in the rest 6 sampling sites (Fig. 2b). The mean total P content was found as 652.31 and 676.54 mg kg^{-1} in topsoil and subsoil, respectively, for Tangail district (Fig. 2b).

The total Cd concentrations in topsoil and subsoil were inconsistent among the sampling sites in the study area, with concentrations ranging from 0.83 to 4.08 mg kg^{-1} in topsoil and from 0.67 to 3.74 mg kg^{-1} in subsoil. Among 10 upazilas, the Cd concentrations were higher in the topsoil than the subsoil in Tangail sadar, Ghatail, Bhuapur, Basail and Delduar upazilas. In other upazilas, the reverse was evident with lower Cd concentrations in topsoil than in the subsoil (Fig. 2c). Higher Cd concentrations were detected at sampling points in Sakhipur (3.14 ± 0.25 mg kg^{-1} in topsoil; 3.18 ± 0.21 mg kg^{-1} in subsoil), Basail (3.51 ± 0.16 mg kg^{-1} in topsoil; 3.37 ± 0.07 mg kg^{-1} in subsoil), Delduar (3.63 ± 0.21 mg kg^{-1} in topsoil; 3.53 ± 0.10 mg kg^{-1} in subsoil) and Mirzapur (3.65 ± 0.29 mg kg^{-1} in topsoil; 3.62 ± 0.12 mgkg^{-1} in subsoil) than other sites. The spatial distribution of Cd in topsoil and subsoil of the study area is presented in Fig. 4, depicting that the upazilas at southern part of Tangail district are highly contaminated with Cd in both topsoil and subsoil. The total Cd concentrations in soils of all upazilas exceeded the environmental quality standard of 1.0 mg kg^{-1} (Fig. 2c). As an overall estimation for Tangail district, the mean total Cd concentration was observed as 2.17 and 2.10 mg kg^{-1} in topsoil and subsoil, respectively, which also exceeded the environmental quality standard (Fig. 2c). The calculated
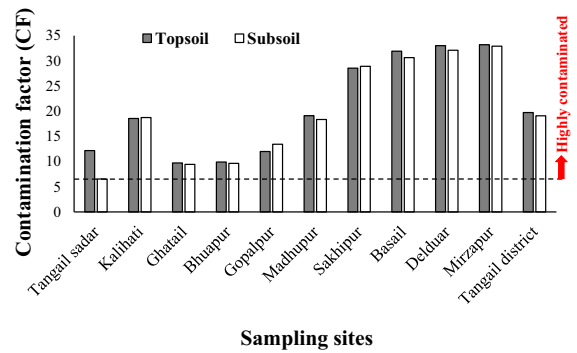

Fig. 3 Level of Cd contamination in sampling sites of Tangail district. The CF values were calculated using 0.11 mg kg^{-1} as the background value for Cd in Bangladesh soil (Kashem and Singh 1999)

values of CF indicated that the topsoil and subsoil of the study area are highly contaminated with Cd (Fig. 3), representing potential ecological and human health risks in the study area. Although high Cd contamination was observed in the study area, especially in the southern part of Tangail district, a considerable amount of areas having Cd concentrations below the permissible levels (1.0 mg kg^{-1}) were also identified (Fig. 4).

Effects of organic amendments on crop production and p uptake

Application of biochar and vermicompost in Cd-contaminated soil, individually or in combination with each other or with TSP fertilizer, significantly ($p < 0.05$) increased plant height, fresh weight and dry weight biomass, fresh yield and P uptake by *A. cruentus* in experimental pots when compared with control treatment (Figs. 5, 6 and Table S2). It was observed that the application of biochar with TSP (T14: 10 t ha^{-1} + 80 kg ha^{-1}) maximized the plant height of by 180%, while the biochar in combination with vermicompost (T11: 5 t ha^{-1} + 5 t ha^{-1}) increased plant height by 161% (Table S2). In the case of only biochar, application of 1, 5 and 10 t ha^{-1} increased the plant height by 44, 62 and 131%, respectively, while 20 t ha^{-1} biochar application (T4) slightly reduced the plant height which revealed that the increased rate of biochar application might not be suitable for optimum growth of *A. cruentus*. Increase in plant height was also observed with 5 t ha^{-1}
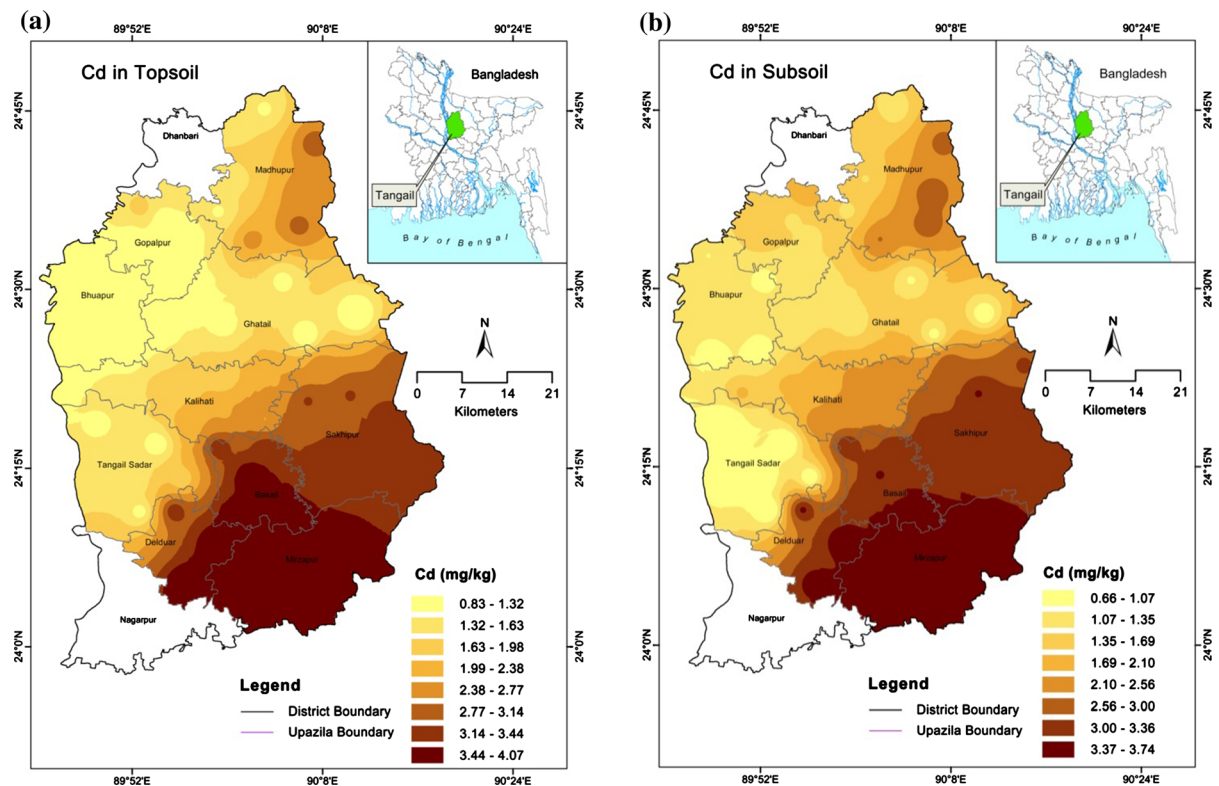
Fig. 4 Spatial distribution of total Cd concentrations (mg kg^{-1}) in topsoil **a** and subsoil **b** of different upazilas in Tangail district, Bangladesh

vermicompost treatment (T6) followed by 1 t ha^{-1} (T5) and 10 t ha^{-1} (T7) treatments (Fig. 5a). Nevertheless, the application of 20 t ha^{-1} vermicompost (T8) significantly ($p < 0.05$) increased the height by 170% over the control treatment (Table S2) which was almost similar to the results of maximum plant height obtained by combined treatment of biochar and TSP fertilizer (T14). Moreover, the results showed that the height of *A. cruentus* plants increased with increasing doses of combined treatments of biochar and vermicompost or TSP fertilizer (Fig. 5a).

In terms of plant biomass, the application of 20 t ha^{-1} vermicompost (T8) alone significantly ($p < 0.05$) increased both fresh and dry weight biomass of *A. cruentus* as compared to the control treatment by 576 and 800%, respectively (Table S2). On the other hand, plant fresh weight biomass of was increased by 51, 207, 289% with the biochar treatment at the rate 1, 5 and 10 t ha^{-1}, respectively (Table S2). Contrariwise, 20 t ha^{-1} biochar application (T4) also reduced the above ground fresh and dry weight biomass (Fig. 5b) as it reduced the plant height

(Fig. 5a). When combined treatments were applied, the highest above ground biomass of *A. cruentus* plants was observed with treatment T11 (5 t ha^{-1} biochar + 5 t ha^{-1} vermicompost) which significantly ($p < 0.05$) increased the fresh and dry weight biomass by 441 and 642%, respectively (Table S2). Although integrated treatments of 20 t ha^{-1} biochar and 80 kg ha^{-1} TSP (T14) significantly increased the biomass of *A. cruentus* as compared to control treatment, this treatment was found to be less effective than the treatment T11.

Moreover, the effectiveness of various single and combined treatments for increasing yield was almost similar with those found for increasing fresh weight biomass since the fresh yield was calculated from the above ground fresh weight biomass of *A. cruentus*. Therefore, application of 20 t ha^{-1} vermicompost (T8) alone was found to be the best treatment which increased the yield of *A. cruentus* almost seven times than that of control treatment. With only biochar treatments, the fresh yield of *A. cruentus* was also increased when biochar was applied at a rate of 1–10
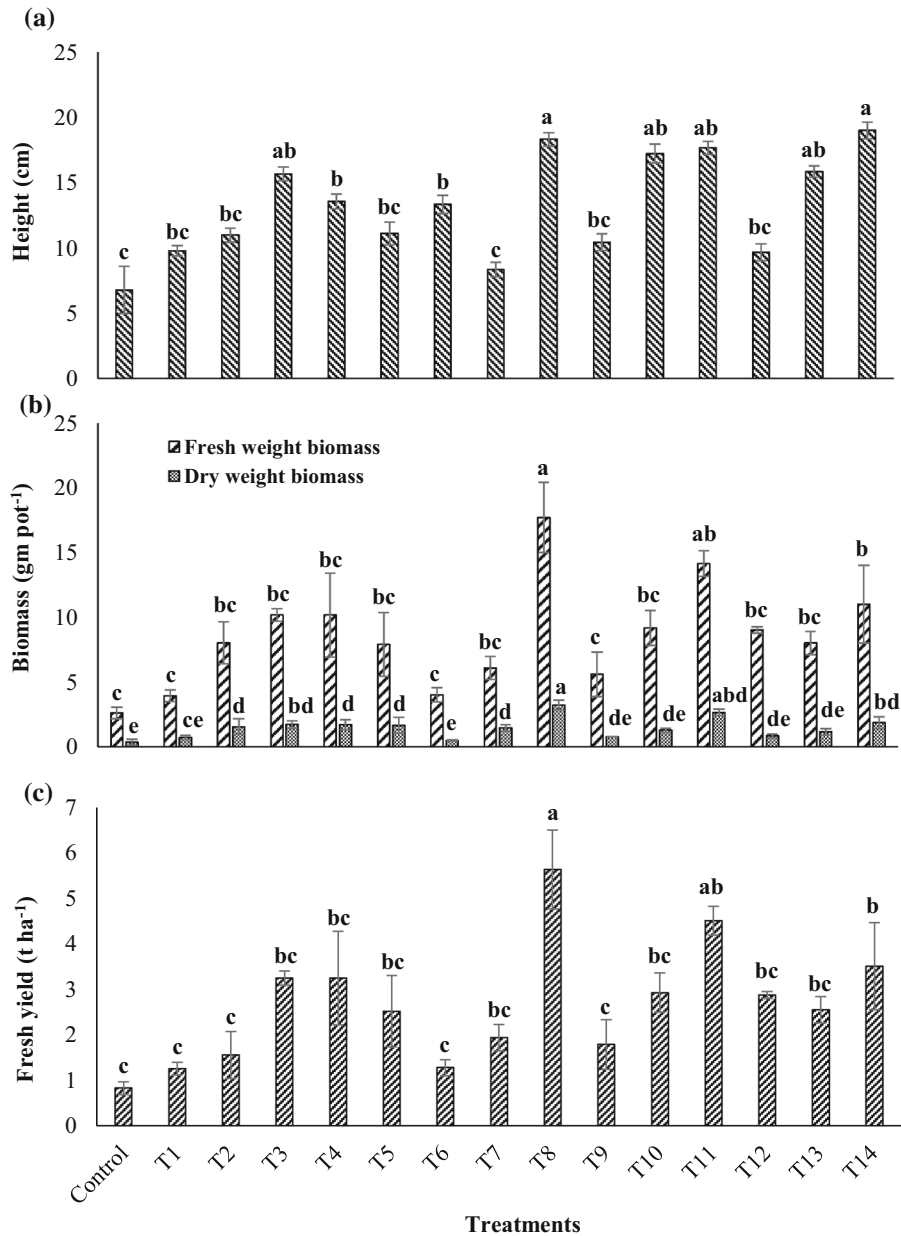
Fig. 5 Effects of different treatments of organic amendments with or without TSP on the height **a**, above ground fresh and dry weight biomass **b**, and yield **c** of *A. cruentus*. Here, Control = Without amendment, T1 = Biochar 1 t ha^{-1}, T2 = Biochar 5 t ha^{-1}, T3 = Biochar 10 t ha^{-1}, T4 = Biochar 20 t ha^{-1}, T5 = Vermicompost 1 t ha^{-1}, T6 = Vermicompost 5 t ha^{-1}, T7 = Vermicompost 10 t ha^{-1}, T8 = Vermicompost 20 t ha^{-1}, T9 = Biochar 0.5 t ha^{-1} + Vermicompost 0.5 t ha^{-1}, T10 = Biochar 2.5 t ha^{-1} + Vermicompost 2.5 t ha^{-1}, T11 = Biochar 5 t ha^{-1} + Vermicompost 5 t ha^{-1}, T12 = Biochar 1 t ha^{-1} + TSP 40 kg ha^{-1}, T13 = Biochar 5 t ha^{-1} + TSP 60 kg ha^{-1}, and T14 = Biochar 10 t ha^{-1} + TSP 80 kg ha^{-1}. The data represent the mean of three replicates and the error bars are standard errors, while the columns denoted by different lowercase letters (a, b, c, d, e) indicate significant ($p < 0.05$) differences among the plant growth and yield parameters due to application of different treatments

t ha^{-1}, while yield was reduced when 20 t ha^{-1} biochar (T4) was applied (Fig. 5c). While among the combined treatments, the highest fresh yield was observed when 5 t ha^{-1} biochar was applied in combination with 5 t ha^{-1} vermicompost (T11) followed by combined treatment of biochar (10 t
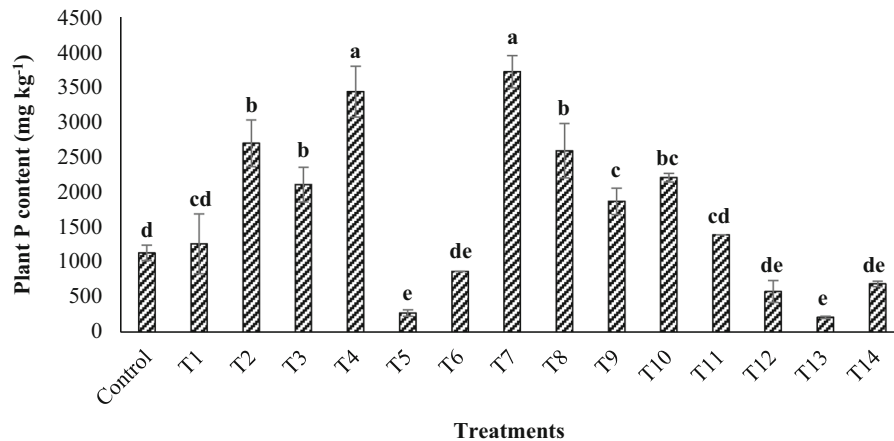
Fig. 6 Effects of different treatments of organic amendments with or without TSP on the total P content of *A. cruentus*. Here, Control = Without amendment, T1 = Biochar 1 t ha^{-1}, T2 = Biochar 5 t ha^{-1}, T3 = Biochar 10 t ha^{-1}, T4 = Biochar 20 t ha^{-1}, T5 = Vermicompost 1 t ha^{-1}, T6 = Vermicompost 5 t ha^{-1}, T7 = Vermicompost 10 t ha^{-1}, T8 = Vermicompost 20 t ha^{-1}, T9 = Biochar 0.5 t ha^{-1} + Vermicompost 0.5 t ha^{-1}, T10 = Biochar 2.5 t ha^{-1} + Vermicompost 2.5 t ha^{-1}, T11 = Biochar 5 t ha^{-1} + Vermicompost 5 t ha^{-1}, T12 = Biochar 1 t ha^{-1} + TSP 40 kg ha^{-1}, T13 = Biochar 5 t ha^{-1} + TSP 60 kg ha^{-1}, and T14 = Biochar 10 t ha^{-1} + TSP 80 kg ha^{-1}. The data represent the mean of three replicates and the error bars are standard errors, while the columns denoted by different lowercase letters (a, b, c, d, e) indicate significant ($p < 0.05$) differences among the plant P content due to application of different treatments

ha^{-1}) and TSP (80 kg ha^{-1}). It can, therefore, be highlighted that higher levels of vermicompost application increased the growth and yield of *A. cruentus*. However, combined application of moderate doses of biochar and vermicompost was found to be the best treatment for increased and sustainable crop production.

The study also found that lower doses of vermicompost or combined application of biochar and TSP significantly reduced the plant P content (Fig. 6). Application of higher doses of biochar (T4) was found to be the best treatment for increased P uptake by *A. cruentus* which increased total P content by 204% over the control treatment (Table S2). The study revealed that P uptake by *A. cruentus* was increased with increasing doses of biochar though slightly different result was obtained by 10 t ha^{-1} biochar treatment (T3). On the other hand, lower doses of vermicompost (1–5 t ha^{-1}) reduced the plant total P content, while maximum increase in P content was found in the case of 10 t ha^{-1} vermicompost application (T7) though 20 t ha^{-1} vermicompost (T8) application slightly reduced the amount as compared to T7 (Table S2). Although total P content was increased when biochar (5 t ha^{-1}) was applied in combination with vermicompost (5 t ha^{-1}); however, the treatment was less effective than

the single treatments of biochar or vermicompost (Fig. 6).

Effects of organic amendments on heavy metals uptake by plant

In this study, all the treatments excluding T5 (vermicompost 1 t ha^{-1}) and T10 (biochar 2.5 t ha^{-1} + vermicompost 2.5 t ha^{-1}) significantly ($p < 0.05$) reduced the Cd uptake in *A. cruentus* as compared to the control (Fig. 7a). The treatments decreased Cd concentrations in *A. cruentus* in the order of: T1 > T13 > T8 = T11 ∼ T2 = T3 > T7 ∼ T4 > T9 = T14 ∼ T12 > T6 > T10 > T5 > Control. The lowest Cd concentration was measured in plants grown in soils treated by 1 t ha^{-1} biochar (T1) which decreased Cd uptake by 78% over the control treatment (Table S2). The study, therefore, revealed that application of biochar (5 t ha^{-1}) in combination with vermicompost (5 t ha^{-1}) is also found to be the best amendment for immobilizing Cd in soils and decreasing uptake by plants.

Since the experimental soil also contaminated with Cr and Pb (Table S1), effects of various treatments on plant uptake of Cr and Pb were also investigated. The results exhibited that application of biochar and vermicompost, alone or in combination with each
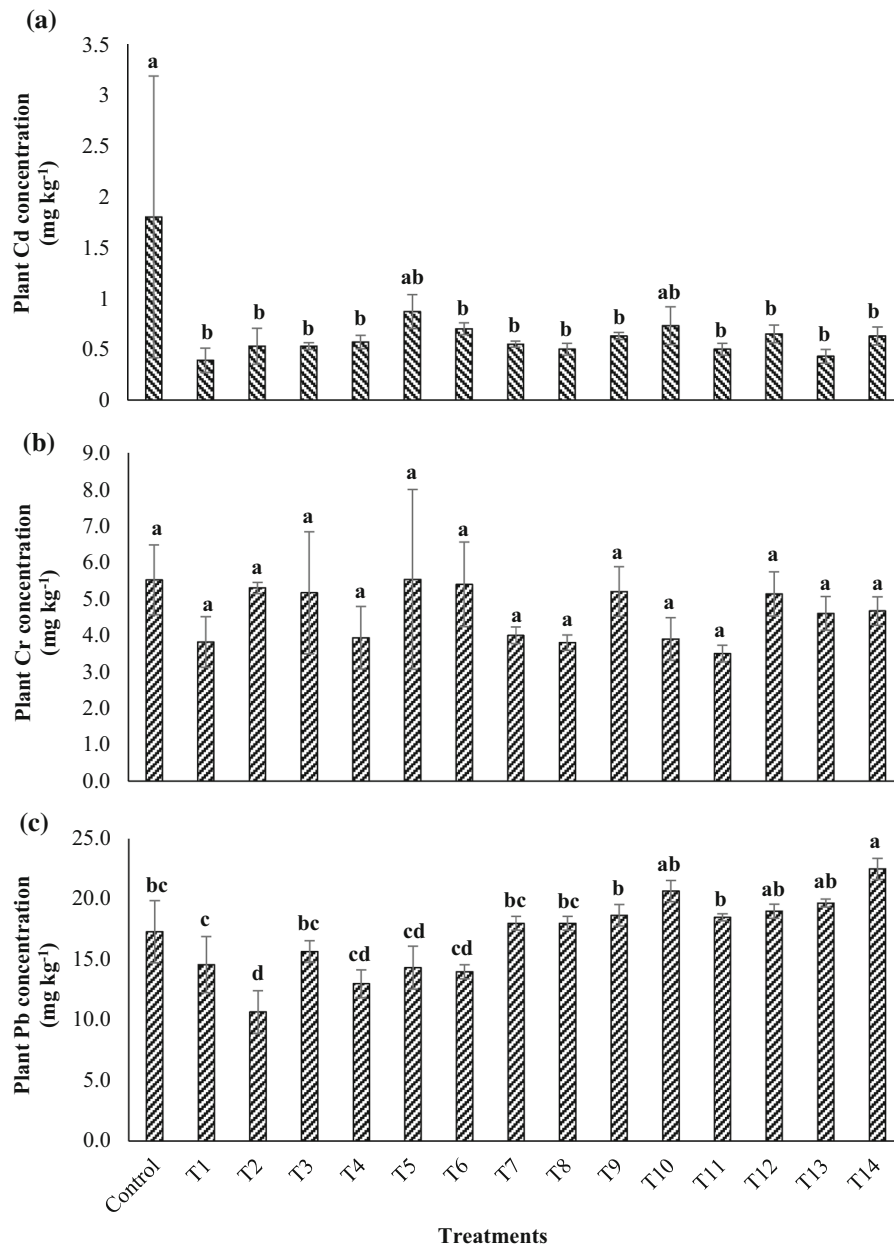
Fig. 7 Effects of different treatments of organic amendments with or without TSP on the uptake of Cd **a**, Cr **b**, and Pb **c** by *A. cruentus*. Here, Control = Without amendment, T1 = Biochar 1 t ha^{-1}, T2 = Biochar 5 t ha^{-1}, T3 = Biochar 10 t ha^{-1}, T4 = Biochar 20 t ha^{-1}, T5 = Vermicompost 1 t ha^{-1}, T6 = Vermicompost 5 t ha^{-1}, T7 = Vermicompost 10 t ha^{-1}, T8 = Vermicompost 20 t ha^{-1}, T9 = Biochar 0.5 t ha^{-1} + Vermicompost 0.5 t ha^{-1}, T10 = Biochar 2.5 t ha^{-1} + Vermicompost 2.5 t ha^{-1}, T11 = Biochar 5 t ha^{-1} + Vermicompost 5 t ha^{-1}, T12 = Biochar 1 t ha^{-1} + TSP 40 kg ha^{-1}, T13 = Biochar 5 t ha^{-1} + TSP 60 kg ha^{-1}, and T14 = Biochar 10 t ha^{-1} + TSP 80 kg ha^{-1}. The data represent the mean of three replicates and the error bars are standard errors, while the columns denoted by different lowercase letters (a, b, c, d) indicate significant ($p < 0.05$) differences among the plant heavy metal contents owing to application of different treatments

other or with TSP, did not significantly reduce plant Cr uptake and Cr concentrations in *A. cruentus* (Fig. 7b). The lowest Cr concentration was detected in the pot amended with 5 t ha^{-1} biochar along with 5 t ha^{-1} vermicompost (T11) which reduced the Cr uptake by 37% over the control treatment (Table S2). In the case of Pb, biochar treatment at a rate of 5 t ha^{-1} significantly reduced the concentration in *A. cruentus.* No significant effect was observed for other treatments, except for T14 (biochar 10 t ha^{-1} + TSP 80 kg ha^{-1}) which significantly increased the Pb concentration in plants (Fig. 7c).

Relationships among organic amendment application, plant growth and heavy metal uptake

Pearson's correlation analysis was performed to determine the relationships among organic amendments, plant growth parameters, P contents as well as heavy metal (Cd, Cr and Pb) concentrations of *A. cruentus* which is shown in Fig. 8. The calculated values of Pearson's correlation coefficients showed a statistically significant strong positive correlation among plant growth parameters, i.e., height-fresh weight biomass (r = 0.764**), height-dry weight biomass (r = 0.672**), and height-fresh yield (r = 0.764**). Moreover, organic amendments showed significantly strong positive correlation with plant growth parameters of (r = 0.569* for plant height, r = 0.725** for fresh weight biomass, r = 0.751** for dry weight biomass, and r = 0.725** for fresh yield), and with P contents (r = 0.599*). On the other hand, organic amendments showed no significant correlation with plant Cd and Pb contents, while significantly strong negative correlation was observed between organic amendment application and Cr uptake by *A. cruentus* (r = − 0.566*). However, no significant association was observed among studied heavy metals (Fig. 8). The heat map of Pearson's correlation clearly depicts that the application of organic amendments, alone or in combination, significantly increases the growth and yield of *A. cruentus* as well as plant P uptake, while reduces heavy metal (Cd, Cr and Pb) uptake resulting from immobilization of

	Height	Fresh weight biomass	Dry weight biomass	Fresh yield	P content	Cd concentration	Cr concentration	Pb concentration	Organic amendments
Height	1.000								
Fresh weight biomass	** 0.764	1.000							
Dry weight biomass	** 0.672	** 0.937	1.000						
Fresh yield	** 0.764	** 1.000	** 0.937	1.000					
P content	-0.057	0.197	0.314	0.197	1.000				
Cd concentration	-0.449	-0.431	-0.417	-0.431	-0.211	1.000			
Cr concentration	-0.419	-0.493	* -0.522	-0.493	-0.428	0.477	1.000		
Pb concentration	0.400	0.253	0.109	0.253	-0.279	0.036	-0.247	1.000	
Organic amendments	* 0.569	** 0.725	** 0.751	** 0.725	* 0.599	-0.376	* -0.566	-0.024	1.000

Fig. 8 Heat map of Pearson's correlation among organic amendment application, various growth parameters, P and heavy metals (Cd, Cr and Pb) concentrations of Red amaranth. Double asterisks sign (**) indicates the correlation is significant at the 0.01 level (2-tailed), while single asterisk sign (*) represents the correlation is significant at the 0.05 level (2-tailed)

heavy metals in contaminated soil by organic amendments.

Human health risk assessment

The human health risk assessment was performed considering both adults and children to evaluate risk factors for causing probable non-carcinogenic and carcinogenic health hazards through the dietary uptake of *A. cruentus* contaminated with Cd, Cr and Pb.

Estimated daily intake (EDI) of heavy metals

The EDIs of Cd, Cr and Pb through the consumption of vegetables were evaluated by considering only the edible part and according to the mean concentration of each metal under different treatments. The EDIs of Cd, Cr and Pb for both adults and children were higher for the vegetables collected from the pots with control treatment. However, the EDIs of all the metals were less than the maximum tolerable daily intake (MTDI) indicating that the vegetables are safe for dietary intake (Table 2). The results confirmed that the application of organic amendments, individually and in combination, decreased the EDI of heavy metals.

Non-carcinogenic and carcinogenic risk of heavy metals

The target hazard quotient (THQ) and hazard index (HI) for non-carcinogenic risk are presented in Table 3, whereas the target carcinogenic risk (TR) values for Cd, Cr and Pb are shown in Table 4. The THQ values of Cd, Cr and Pb were < 1 for all the treatments, while the HI values were also < 1 for both adults and children (Table 3), indicating that no potential non-carcinogenic risk. On the other hand, the TR values of Cd showed there is no carcinogenic risk for adults and children due to consumption of vegetables grown with different treatments (Table 4). In most of the cases, the TR values of Pb were higher than the acceptable limit of 1.0E-04 (USEPA 1989) which exhibited high carcinogenic risk for both adults and children.

Discussion

Properties of agricultural soils and level of cadmium (Cd) contamination

Detailed information about soil pH is important in relation to agricultural practices since pH regulates

Table 2 Estimated daily intake (EDI) of heavy metals by adults and children through consumption of contaminated red amaranth (*A. cruentus*) and corresponding maximum tolerable daily intake (MTDI)

| Treatments | EDI (mg person^{-1} day^{-1}) of heavy metals | | | | | |
| | Adults | | | Children | | |
	Cd	Cr	Pb	Cd	Cr	Pb
Control	0.0011	0.0033	0.0103	0.0027	0.0084	0.0264
T1	0.0002	0.0018	0.0068	0.0005	0.0046	0.0174
T2	0.0003	0.0027	0.0055	0.0007	0.0070	0.0141
T3	0.0002	0.0023	0.0069	0.0006	0.0058	0.0176
T4	0.0003	0.0017	0.0057	0.0006	0.0044	0.0146
T5	0.0005	0.0030	0.0078	0.0012	0.0077	0.0199
T6	0.0002	0.0017	0.0043	0.0006	0.0043	0.0111
T7	0.0003	0.0025	0.0112	0.0009	0.0064	0.0286
T8	0.0002	0.0018	0.0084	0.0006	0.0045	0.0215
T9	0.0002	0.0016	0.0058	0.0005	0.0041	0.0148
T10	0.0003	0.0014	0.0075	0.0007	0.0036	0.0192
T11	0.0002	0.0017	0.0091	0.0006	0.0044	0.0233
T12	0.0002	0.0013	0.0049	0.0004	0.0034	0.0126
T13	0.0002	0.0018	0.0076	0.0004	0.0046	0.0195
T14	0.0003	0.0020	0.0099	0.0007	0.0053	0.0253
MTDI	0.046[a]	0.2[b]	0.21[a]	0.046[a]	0.2[b]	0.21[a]

[a]JECFA (2003), [b]RDA (1989)

Table 3 Target hazard quotient (THQ) and hazard index (HI) of heavy metals in red amaranth (*A. cruentus*) for adults and children

Treatments	THQ						HI	
	Adults			Children			Adults	Children
	Cd	Cr	Pb	Cd	Cr	Pb		
Control	0.0021	0.0066	0.0206	0.0055	0.0168	0.0527	0.0293	0.0750
T1	0.0004	0.0035	0.0136	0.0009	0.0091	0.0348	0.0175	0.0448
T2	0.0005	0.0055	0.0110	0.0014	0.0140	0.0283	0.0170	0.0437
T3	0.0005	0.0045	0.0138	0.0012	0.0116	0.0353	0.0188	0.0481
T4	0.0005	0.0035	0.0114	0.0013	0.0089	0.0293	0.0154	0.0395
T5	0.0009	0.0060	0.0156	0.0024	0.0154	0.0399	0.0225	0.0577
T6	0.0004	0.0033	0.0087	0.0011	0.0086	0.0223	0.0124	0.0320
T7	0.0007	0.0050	0.0223	0.0017	0.0127	0.0572	0.0280	0.0716
T8	0.0005	0.0035	0.0167	0.0012	0.0091	0.0429	0.0207	0.0532
T9	0.0004	0.0032	0.0116	0.0010	0.0083	0.0297	0.0152	0.0390
T10	0.0005	0.0028	0.0149	0.0014	0.0072	0.0383	0.0182	0.0469
T11	0.0005	0.0034	0.0182	0.0013	0.0088	0.0466	0.0221	0.0567
T12	0.0003	0.0027	0.0098	0.0009	0.0068	0.0252	0.0128	0.0329
T13	0.0003	0.0036	0.0152	0.0009	0.0091	0.0391	0.0191	0.0491
T14	0.0006	0.0041	0.0198	0.0014	0.0105	0.0507	0.0245	0.0626

Table 4 Target carcinogenic risk (TR) of heavy metals via consumption of contaminated red amaranth (*A. cruentus*)

Treatments	TR					
	Adults			Children		
	Cd	Cr	Pb	Cd	Cr	Pb
Control	1.6E−05	4.9E−05	**1.54E−04**	4.1E−05	**1.3E−04**	**4.0E−04**
T1	3.0E−06	2.7E−05	**1.02E−04**	7.0E−06	7.0E−05	**2.6E−04**
T2	4.0E−06	4.1E−05	8.30E−05	1.1E−05	**1.1E−04**	**2.1E−04**
T3	3.0E−06	3.4E−05	**1.03E-04**	9.0E−06	9.0E−05	**2.6E−04**
T4	4.0E−06	2.6E−05	8.60E−05	1.0E−05	7.0E−05	**2.2E−04**
T5	7.0E−06	4.5E−05	**1.17E−04**	1.8E−05	**1.2E−04**	**3.0E−04**
T6	3.0E−06	2.5E−05	6.50E−05	8.0E−06	6.0E−05	**1.7E−04**
T7	5.0E−06	3.7E−05	**1.67E−04**	1.3E−05	**1.0E−04**	**4.3E−04**
T8	3.0E−06	2.7E−05	**1.26E−04**	9.0E−06	7.0E−05	**3.2E−04**
T9	3.0E−06	2.4E−05	8.70E−05	8.0E−06	6.0E−05	**2.2E−04**
T10	4.0E−06	2.1E−05	**1.12E−04**	1.0E−05	5.0E−05	**2.9E−04**
T11	4.0E−06	2.6E−05	**1.36E−04**	9.0E−06	7.0E−05	**3.5E−04**
T12	3.0E−06	2.0E−05	7.40E−05	6.0E−06	5.0E−05	**1.9E−04**
T13	2.0E−06	2.7E−05	**1.14E−04**	6.0E−06	7.0E−05	**2.9E−04**
T14	4.0E−06	3.1E−05	**1.48E−04**	1.1E−05	8.0E−05	**3.8E−04**

Bold indicates the value exceeded the acceptable value of 1.0E-04 (USEPA 1989)

plant nutrients availability and influences the activity of soil microorganisms (Kadam 2016; Oshunsanya 2018). The study revealed extremely acidic to slightly alkaline topsoil (4.4–7.7) and slightly acidic to moderately alkaline subsoil (6.2–7.9) in the study area (Fig. 2a). Rahman et al. (2012) reported that the topsoil pH in Madhupur upazila of Tangail district ranged from 4.3 to 6.1 with an average of 5.3. Miah et al. (2016) found relatively lower topsoil pH (4.7–5.6) than the subsoil pH (5.4–6.2) in Basail upazila of Tangail district. Sheel et al. (2015) found lower topsoil pH in Mymensingh (5.4) and Jamalpur

(5.8) districts which are two adjoining districts of Tangail district, whereas the subsoil pH was 6.3 and 6.2, respectively. Thus, the study found almost similar levels of soil pH in Tangail, Mymensingh and Jamalpur districts. Similar results were also reported by Egashira et al. (2003) who observed mean topsoil pH of 5.2, 6.7, 5.1, 7.6, 5.7, 6.3 and 5.0 in Bogra, Rajshahi, Panchagarh, Khulna, Patuakhali, Mymensingh and Comilla districts, respectively, with mean subsoil pH of 7.2, 7.2, 5.4, 8.0, 6.8, 7.5 and 7.0, respectively.

The topsoil acidity in the study area might be attributed to acid rain, increased crop production, and excessive use of fertilizers, mainly nitrogen (N) fertilizers (Miah et al. 2016; Oshunsanya 2018). For instance, H^+ ions released during the nitrification of ammonium (NH_4^+) fertilizers can lead to the topsoil acidity, while rainwater containing acids like tetraoxosulfate and trioxonitrate acid can also result in the topsoil acidity of agricultural lands nearby urban and industrial areas (Oshunsanya 2018). Kumar et al. (2018) observed that the topsoil pH of Tangail district is gradually decreasing and reported that the mean topsoil pH was slightly alkaline before 2000 which now changed to slightly acidic to neutral. However, the optimum pH for most agricultural crops ranges from 5.5 to 7.5 (Oshunsanya 2018), indicating that the soil pH of Tangail district is suitable for agricultural production.

The mean total P content observed in topsoil (652 mg kg^{-1}) and subsoil (676 mg kg^{-1}) of the study area was within the normal P contents in soils ranged from 150 to 700 mg kg^{-1} (Menzies 2009). The results also revealed that the total P in both topsoil and subsoil at different upazilas of Tangail district exceeded the critical level of total P contents (150–700 mg kg^{-1}), even higher concentrations of total P were observed in few sampling points (Fig. 2b). Such a higher total P contents in the topsoil and subsoil might be attributed to the intensive agricultural practices with continuous application of P fertilizers for agricultural production in the study area (Egashira et al. 2003; Andersson et al. 2015). Egashira et al. (2003) also observed similar results for total P concentrations in topsoil and subsoil of other seven districts of Bangladesh. They found that the total P concentration was 203, 411, 604, 561, 417, 268 and 362 mg kg^{-1} in topsoil of Bogra, Rajshahi, Panchagarh, Khulna, Patuakhali, Mymensingh and Comilla

districts, respectively, whereas the concentration was 126, 287, 688, 609, 605, 245 and 472 mg kg^{-1} in subsoil of those districts, respectively (Egashira et al. 2003). In agricultural soils, P is usually accumulated near the surface of the soil, i.e., in topsoil as a result of the frequent inputs of P fertilizers (Menzies 2009), while in subsoil P can be concentrated naturally or also through the repeated application of P fertilizers resulting from higher P leaching from the topsoil (Andersson et al. 2015). Though plants uptake the required amount of P mainly from the topsoil, subsoil also significantly contributes to the supply of P to the plants particularly when P is available early in the growth season (McBeath et al. 2012).

Higher concentrations of Cd were found in the topsoil (2.17 mg kg^{-1}) and subsoil (2.10 mg kg^{-1}) of the study area (Fig. 2c). Since Bangladesh has yet little industrial pollution of agricultural lands (Meharg et al. 2013), agricultural activities using frequent inputs of Cd containing phosphate fertilizers are considered to be the primary causes of Cd contamination in the studied agricultural soils. However, industrial activities are also contributing to soil heavy metal contamination including Cd in Tangail district with the expansion of industrial units and subsequent activities from the nearby industrialized cities like Dhaka and Gazipur (Kormoker et al. 2020). The indiscriminate growth of industries in the southern part of the Tangail region might be the possible cause behind the inconsistent results observed in terms of Cd concentrations in the study area, whereas higher levels of Cd contamination in both topsoil and subsoil at the upazilas located in the southern part of the Tangail district and adjacent to Dhaka and Gazipur districts (Fig. 4). Moreover, the contribution of natural sources to the soil Cd concentrations in the study area cannot be underestimated. Tusher et al. (2017) investigated the Cd concentration in agricultural soils and observed 1.4 ± 1.13 and 1.18 ± 0.42 mg kg^{-1} of Cd in topsoil of Tangail sadar and Mirzapur upazila, respectively. Islam et al. (2018b) also investigated the Cd contamination in agricultural soils in Tangail sadar upazila and found that the topsoil is highly contaminated with Cd, ranging from 0.71 to 3.53 mg kg^{-1}. Moreover, higher total Cd concentrations were also found in soils of other parts in Bangladesh, for example, 3.15 mg kg^{-1} in Chorfasion upazila (Bhola), 2.89 mg kg^{-1} in Kaliganj upazila (Jhenaidah), 5.23 mg kg^{-1} in Ishwardi upazila (Pabna),

2.39 mg kg^{-1} in Lalpur upazila (Natore), 2.40 mg kg^{-1} in Dumuria upazila (Khulna), and 2.43 mg kg^{-1} in Asasuni upazila (Satkhira) (Mamun et al. 2018). Islam et al. (2015a) reported the mean Cd concentration of 6.3 ± 3.8 mg kg^{-1} in agricultural soils (0–20 cm) of Bogra district, Bangladesh. On the other hand, this study reported higher mean Cd concentrations in soils as compared to other countries such as China (Chen et al. 2019; Mo et al. 2020), India (Kaur et al. 2019; Kumar et al. 2019), Egypt (Asmoay et al. 2019), and Morocco (Ennaji et al. 2020).

Effects of organic amendments on crop production and p uptake

The study observed that application of biochar in combination with vermicompost is the best treatment for increased plant growth, yield and P uptake by *A. cruentus*. This might be due to the fact that co-application of vermicompost and biochar amendments have synergistic effects on soil nutrients and microbial community structure which positively influences the plant growth and biomass (Song et al. 2015). Di et al. (2019) revealed that application of biochar combined with low and high doses of vermicompost significantly ($p < 0.05$) increased the plant height and rice yield by 23–39% when compared with that achieved without biochar. This might be attributed to the interactive effects of biochar on the functioning of vermicompost agents (Di et al. 2019). Since the surfaces and pores of biochar provide a habitat for microorganisms, its amendment improves bulk density, air circulation, and the amount of water and nutrients in the soil matrix (Lehmann et al. 2011). These physicochemical properties promote microbial abundance and activities by providing space and expanded niches for vermicompost (Ngo et al. 2013), which results in stronger microbial development and consequently changes insoluble or fixed nutrients to plant-available forms in soil (Song et al. 2015). Though combined application of higher doses of biochar and TSP was found to be efficient to increase plant height, it could not increase crop yield as compared to single or combined treatments using biochar and vermicompost (Fig. 5).

Moreover, combined treatment of biochar and vermicompost significantly increased the P uptake by *A. cruentus* as compared to control treatment though the efficiency was not as much higher as obtained using the biochar alone (Fig. 6). The increase

in soil pH as a result of combined application of biochar and vermicompost also increases the available P in the soils by reducing Fe and Al activity (Mensah and Frimpong 2018), which might be the possible reason of higher P uptake and total P contents in *A. cruentus* plants. Ch'ng et al. (2014) also found that application of biochar and compost, alone or in combination, significantly increased the total P, available P, inorganic P fractions, and organic P. In the case of single application of biochar, higher doses of biochar application (20 t ha^{-1}) reduced the plant height, above ground biomass and P uptake. Similar result was found by Hamzah and Shuhaimi (2017) who reported that increased rate of biochar application to the soil reduced the heights of oil palm and rubber seedlings due to nutrient deficiency resulted from increasing soil pH. Situmeang et al. (2018) also reported that higher levels of bamboo biochar application reduced the height of *Amaranthus tricolor* L. Mohawesh et al. (2018) observed that biochar applications at higher levels adversely affected tomato and bell pepper plant growth, and lower levels of biochar applications resulted in significantly greater biomass production than the higher biochar levels. Such decrease in biomass production at the higher levels of biochar addition was likely the result of a high pH and EC in the biochar treatments which subsequently causes nutrient deficiency as a consequence of increasing of soil pH and EC (Mohawesh et al. 2018). In addition, Piash et al. (2019) reported that farmyard biochar significantly increased the P uptake by *Amaranthus lividus*. However, maximum efficiency was observed when biochar was applied combinedly with inorganic fertilizer (Piash et al. 2019), which was totally different from our findings.

On the contrary, application of vermicompost at higher doses (20 t ha^{-1}) maximized the plant growth and yield (Fig. 5). Hadi et al. (2011) also reported that plant height and yield of *Matricaria chamomomile* L. were increased with increasing doses of vermicompost, while maximum result was obtained when 20 t ha^{-1} vermicompost was applied. Similar result was also found by Alam et al. (2007) who found that vermicompost amendments increased plant heights of potato (*Solanum tuberosum* L.) since vermicompost is a good source of plant micro- and macro-nutrients, particularly NPKS. Di et al. (2019) also observed that rice yield was increased with increasing doses of vermicompost treatments as compared to control

treatment. Shirkhodaei et al. (2014) observed that fresh and dry weight biomass of coriander plant (*Coriandrum sativum* L.) increased with increasing the levels of vermicompost treatments as compared to control treatment. Similar results were also reported by Moghadam et al. (2012) who studied the effects of various vermicompost treatments (0–30%) on the growth and development of *Lilium* Asiatic hybrid var. Navona plant. Amendment of soil with vermicompost leads to increase in water holding capacity, nutrients and minor elements concentrations, which in turn lead to a better plant growth and development (Atiyeh et al. 2002). Chaoui et al. (2003) found that application of vermicompost amendment significantly increased the availability of plant essential nutrients, which also supports our findings. Nevertheless, growth of plants can be different in different vermicompost treatments because growth promoting substances are released at different rates in different treatments (Singh et al. 2008; Azarmi et al. 2009).

Several studies (e.g., Pramanik 2010; Jauquet et al. 2011; Song et al. 2015; Goswami et al. 2017) have supported the results of our study as they also reported that higher doses of vermicompost amendments can enhance the soil physicochemical properties, supply sufficient amount of plant-available nutrients, improve soil biological activities, and thereby increase the crop growth and yield. However, vermicompost application at high levels can significantly ($p < 0.01$) increase the cumulative ammonia (NH_3) volatilization and nitrous oxide (N_2O) emissions (Di et al. 2019), resulting in adverse environmental impacts including nitrogen (N) loss and global warming (Arancon et al. 2006). On the other hand, the incorporation of biochar and vermicompost significantly ($p < 0.01$) reduces the cumulative N_2O emission and gaseous N loss (Di et al. 2019). Wang et al. (2019) reported that the combined application of biochar and compost delivers considerable benefits to crop growth and nutrient cycling. Therefore, our study suggests that combined amendment of moderate levels of biochar (5 t ha^{-1}) and vermicompost (5 t ha^{-1}) is the best treatment for optimum crop production while reducing the environmental risk.

Effects of organic amendments on heavy metals uptake by plant

Application of biochar and vermicompost, alone or in combination with each other or with TSP, significantly ($p < 0.05$) reduced plant Cd uptake and decreased the Cd concentrations in *A. cruentus* as compared to control treatment (Fig. 7a). These results are consistent with the findings of other similar experiments conducted with organic amendments where composts and lignite reduced the accumulation of Cd in leafy vegetables and potato (Simmler et al. 2013, Mamun et al. 2016, 2017). The Cd concentration in *A. cruentus* ranged from 0.39 to 1.8 mg kg^{-1} DW which is similar to the concentrations of Cd found in spinach (0.559–1.40 mg kg^{-1} DW) by Naser et al. (2009) in Bangladesh soils although the maximum concentration of Cd in *A. cruentus* is higher than the concentration of Cd in Spinach (*Lactuca sativa*). The high reduction in Cd concentration in *A. cruentus* due to the application of different organic amendments might be due to the small amount of organic matter in the control soils. When the organic matters were added to soils, those made the Cd less available in soil solution and plants could not accumulate. Moreover, the soils textural class was loamy sand in our experiment. Sand has the least CEC compared to silt, clay and organic matters in soils. Similarly, organic matters have higher CEC than soils (Mamun et al. 2016, 2017). Probably, organic matter addition in soils increased the total CEC in soils (although we have not tested the CEC in our soils) that caused less Cd in soil solution and subsequently less accumulation by plants.

Vermicompost applied at the rate of 20 t ha^{-1} reduced the Cd uptake by 72% which was similar to that obtained by combined treatment of biochar (5 t ha^{-1}) and vermicompost (5 t ha^{-1}). Adiloğlu et al. (2017) studied the effect of various doses of vermicompost application on heavy metal contents of cucumber (*Cucumis sativus* L.) and found that Cd contents of cucumber were decreased with increasing the doses of vermicompost application. Since higher levels of vermicompost application are discouraged owing to its adverse environmental impacts (Di et al. 2019), the study recommends that application of biochar (5 t ha^{-1}) in combination with vermicompost (5 t ha^{-1}) is the best treatment for immobilizing Cd in soils and decreasing subsequent uptake by plants while

maximizing crop production without causing additional environmental problems.

Application of organic amendments, individually or in combination, immobilizes or stabilizes Cd in soil by forming stable metal–organic complexes (Ahmad et al. 2015) and Cd immobilization in soil is one of the mechanisms for increased plant biomass in metal-contaminated soil (Dourado et al. 2013). These might be the possible reason by which the combined treatment of moderate doses of biochar and vermicompost (T11) restricted Cd uptake in aerial part of A. cruentus plants along with increased growth and yield. Moreover, the organic amendments transform the existing bioavailable metal species present in soil into geochemically more stable forms whether applied singly or combined (Ahmad et al. 2014). Our findings are in line with the results of Branzini and Zubillaga (2012) who reported that organic amendment was more effective in the immobilization of heavy metals as compared to the combined application of organic amendment and phosphate fertilizer, which might be the probable cause for reduced Cd and Cr uptake by A. cruentus treated with combined treatment of moderate doses of biochar and vermicompost (T11).

From our results, it is observed that although the concentrations of Cr and Pb in soil were low, the plant uptake of Cr and Pb was high indicating the availability of the metals in soil solution which might be due to the nature of our soil that had more than 84% sand and 1.2% organic matter (OM) (multiplied %C by Van Bemmelen factor, 1.724). Application of only biochar regardless of the doses reduced all the studied heavy metals (Cd, Cr and Pb) uptake by A. cruentus although some of them were statistically not significant, indicating that biochar has higher affinity to the metals. On the contrary, maximum Pb content was detected in plants amended with combined treatment of biochar (10 t ha^{-1}) and TSP (80 kg ha^{-1}) which significantly ($p < 0.05$) increased Pb concentration by 30% over the control treatment. Several studies (e.g., Castaldi et al. 2005; Angelova et al. 2010; Houben et al. 2013; Karer et al. 2015; Yin et al. 2016; Li and Gao 2019) found that application of organic amendments, alone or in combination, reduced the bioavailability of Pb. Unlike to these studies, our study found slightly different results which might be due to the increase in Pb mobility resulting from the application of higher doses of biochar with vermicompost. These contradictory findings can be explained partially if the

dynamics of metal-dissolved OM complexes in soil solution are considered (Murray et al. 2011). Plaza et al. (2006) reported that fulvic acid (FA), unlike humic acid (HA), has the ability to increase the bioavailability of metalsin soil. If the organic matter applied as soil amendment is not fully decomposed in the soil, the FA is increased in soils which subsequently enhance the plant metal uptake, for example Pb (Murray et al. 2011).

The application of biochar and vermicompost in soil will not only reduce the accumulation of Cd in plants but also will help a country to progress environmentally and economically. At present, in Bangladesh, the sawdust is thrown in water bodies, as farmers directly cannot use sawdust in agricultural lands. The deposition of sawdust in aquatic bodies is extremely bad for our ground water, surface water and aquatic animals. The conversion of sawdust into biochar and application of biochar in agricultural lands will save the aquatic bodies, aquatic lives and overall environment. In this way the country can increase agricultural production, trap carbon in soils and can save earn money from increased agricultural production and other associated benefits. The increased production of agricultural products due to the application of biochar and vermicompost could help the economy of Bangladesh. However, further studies could be conducted to determine the period of the effects of biochar and vermicompost in agricultural soils to reduce the transfer of Cd from soil to plants, to increase plant growth, to increase organic matter content in soils and to trap carbon in soils. Further study is also warranted to determine the effects of different rates of biochar and vermicompost on the transfer of Cd from different textured soils to different plants.

Health risk assessment

The concentrations of toxic metals (Cd, Cr and Pb) in A. cruentus grown under different treatments were found to be high, indicating probable carcinogenic and non-carcinogenic health risks to the consumers and, thus, preliminary human health risk assessment was performed (Tables 2, 3, 4). The estimation exhibited that application of organic amendments, individually and in combination, decreased the EDI of heavy metals with no non-carcinogenic risk. Moreover, the results indicated that the risk factor was decreased

when some specific treatments with organic amendments were applied. The study thus revealed that application of organic amendments, individually or in combination, reduced the carcinogenic and non-carcinogenic risks of Cd. However, there is a possibility of lifetime cancer risk from Cr and Pb, especially for children. For instance, higher EDI value for Pb was observed with the treatment T7 (vermicompost 10 t ha^{-1}) as a result of higher amount of Pb uptake by the plant biomass under this treatment (Table 2), indicating adverse health implications of vermicompost as a single treatment for both adults and children.

Conclusions

Bangladesh has soils having high Cd concentrations in both topsoil and subsoil along with the co-contamination of other heavy metals. In order to produce safe agricultural products in such Cd-contaminated agricultural soils, our study recommends the application of 5 t ha^{-1} of biochar in combination with 5 t ha^{-1} vermicompost to increase production and to reduce human health risks, particularly caused by Cd and Cr. Although higher doses of vermicompost (20 t ha^{-1}) were also found to be effective for optimum plant growth, increased P uptake and reduced Cd and Cr uptake as a single treatment, such higher level of vermicompost application is known to have potential to cause adverse environmental impacts. The findings of our research will improve our understanding about the effects of individual or combined application of organic amendments on crop production in Cd-contaminated soils with possible co-contamination of Cr and Pb. Furthermore, the results will encourage farmers to extensively use organic amendments instead of repeated use of inorganic phosphate fertilizers which would ultimately reduce the reliance on inorganic fertilizers produced from scarce phosphate rocks. Moreover, future work is required to determine the longevity of the effects of biochar and vermicompost in loamy sand soils in field trials with other crop varieties.

Author contributions SAM was involved in conceptualization, methodology, validation, supervision, writing—reviewing and editing, project administration, funding acquisition; SS was involved in methodology, validation, formal analysis, investigation, data curation, visualization, software, writing—Original draft preparation; JF was involved in formal analysis, investigation, data curation, writing—original draft preparation; TRT was involved in conceptualization, methodology, validation, visualization, software, writing—reviewing and editing; AS was involved in formal analysis, investigation, data curation, writing—original draft preparation; FA was involved in formal analysis, investigation, data curation; MRB was involved in writing—reviewing and editing; ZP was involved in conceptualization, methodology, funding acquisition, writing–reviewing and editing.

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