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Effects of Organic Amendment on Heavy Metal and Macronutrient Contents in Paddy Soil

(Kesan Bahan Pembaik Pulih Organik pada Logam Berat dan Kandungan Makronutrien di dalam Tanih Padi)

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ABSTRACT

The enormous use of chemical fertilizers recently has contributed to negative impacts on soil quality for agriculture purposes. However, if we use the fertilizer along with the organic amendment, it would offer a good approach for sustainable agriculture practice. Therefore, this study examined the effects of HT Organic Compound (HTOC) application on heavy metal and macronutrient content of the soil in a 2-season cultivated paddy field from 2016 to 2017. The sampling period was carried out after the harvest period and this study included three treatments: Chemical fertilizer or NPK (CF), chemical fertilizer and 250 kg organic amendment (CF+HTOC250), and chemical fertilizer and 500 kg organic amendment (CF+HTOC500). Several soil parameters were analyzed for this study such as soil organic matter content, pH, available P, available K, total N, and some heavy metals like As, Cd, Cr, Cu, Pb, and Zn. The results showed that the treated soil (CF+HTOC treatment) showed significantly higher soil organic matter content, pH, available P, available K, and total N than the chemical fertilizer treatment (CF). In the long-term application, the treated soils with HTOC tended to have lower Cd, Cr, Cu and Pb concentrations as compared with the chemical fertilizer treatment (CF). As a whole, this study concluded the application of chemical fertilizer along with HTOC could be an alternative method to improving soil quality in a paddy field area.

Keywords: Chemical fertilizer; heavy metal; HTOC; paddy soil; soil nutrient

ABSTRAK

Penggunaan baja kimia dengan jumlah yang banyak pada masa kini telah menyumbang kepada kesan-kesan negatif terhadap kualiti tanah bagi tujuan pertanian. Tetapi, jika kita menggunakan baja kimia bersama dengan bahan pembaik pulih organik, ia akan menawarkan pendekatan yang baik untuk amalan pertanian lestari. Oleh itu, kajian ini mengkaji kesan penggunaan HT Organic Compound *(HTOC) ke atas logam berat dan kandungan makronutrien tanih selama dua musim penanaman padi dari tahun 2016 hingga 2017. Masa aktiviti persampelan dilakukan selepas masa tuai dan dalam kajian ini terdapat tiga jenis rawatan yang dikaji; baja kimia atau NPK (CF), baja kimia dan 250 kg bahan pembaik pulih organik (CF+HTOC250), baja kimia dan 500 kg bahan pembaik pulih organik (CF+HTOC500). Beberapa paramater tanih yang dianalisis dalam kajian ini seperti kandungan bahan organik tanih, pH, P tersedia, K tersedia, jumlah N dan beberapa unsur logam berat seperti As, Cd, Cr, Cu, Pb dan Zn. Hasil kajian mendapati bahawa tanah yang dirawat (CF+HTOC) menunjukkan kandungan bahan organik tanih, pH dan P tersedia, K tersedia dan jumlah N yang signifikan lebih tinggi daripada rawatan baja kimia (CF). Bagi penggunaan jangka panjang, tanah yang dirawat dengan HTOC cenderung mempunyai kandungan logam berat yang lebih rendah berbanding dengan rawatan baja kimia (CF). Secara keseluruhan, kajian ini menyimpulkan penggunaan baja kimia bersama dengan HTOC boleh menjadi kaedah alternatif untuk membaikpulih kualiti tanih di kawasan sawah padi.*

Kata kunci: Baja kimia; HTOC; logam berat; nutrien tanih; tanih sawah

INTRODUCTION

Chemical fertilizers have been widely used to increase soil fertility and crop production in agricultural activities all over the world (Bhatt et al. 2019). However, the excessive use of the fertilizers had bad impacts on agricultural soils such as the high concentration of radionuclides and heavy metal, soil structural deterioration, soil acidity and others (Sodango et al. 2018). Paddy soil is one of the agricultural soils that is easily degraded due to the intensive use of land. Many studies have highlighted the use of organic amendments to mitigate soil quality issues (Cesarano et al. 2017; Lwin et al. 2018). The organic amendments were found better than chemical fertilizer in terms of physicochemical properties. This was because they have been enriched with nutrient and organic matter contents. The organic amendments could improve the physical properties of soil such as soil density, soil permeability, soil aggregates, microbial and biological function (Meena et al. 2020; Paradelo et al. 2019). However, the application of organic amendments in the field may not fulfill the nutrient requirement for a crop, as it is a 'slow release' which lead to a nutrient deficiency at the first application (Hagemann et al. 2017). Hence, the combined application of organic amendment and chemical fertilizer may draw a good solution for nutrient management. The use of chemical fertilizer along with organic amendment could promote soil microbial activities and soil nutrients in a subtropical paddy field (Li et al. 2018), enhance soil organic matter, macronutrient content and enzyme activity (Ning et al. 2017). Naher et al. (2019) stated nutrient mineralization was affected by fertilizer management practice and temperature. On the whole, the integration of organic amendment can improve the efficiency of chemical fertilizer utilization.

To date, previous studies have applied the organic amendment for amending soil properties such as animal manures, biosolids, compost, organic fertilizer, biochar, and humic acid (Brtnicky et al. 2019; Urra et al. 2019). Unlike the previous studies, we used the combination of organic and mineral components such as humic acid, rubber bark dust, clinoptilolite, and kieserite minerals to make a soil amendment namely HT Organic Compound (HTOC). The mineral content in HTOC provided more additional sources of nutrients as compared with other organic amendments. In this study, the field trial was carried out in continuous 2-seasons paddy cultivation to analyze the effects of HTOC on heavy metal and macronutrient content in paddy soil.

MATERIALS AND METHODS

STUDY SITE

The study site is located at the paddy cultivation area of Alor Senibong, Kedah, Malaysia. In this trial, rice seedlings (MR 219) were planted in each of the field plots with a size of 40 × 40 m^2. A 2-season field trial was carried out, starting from 2016 to 2017, to investigate the potency of HTOC as a source of nutrients in paddy soil. There are three treatments applied as depicted in Table 3. Each treatment plot was split by small drains to preclude transboundary movement of the applied materials (Figure 1). This area had a tropical climate

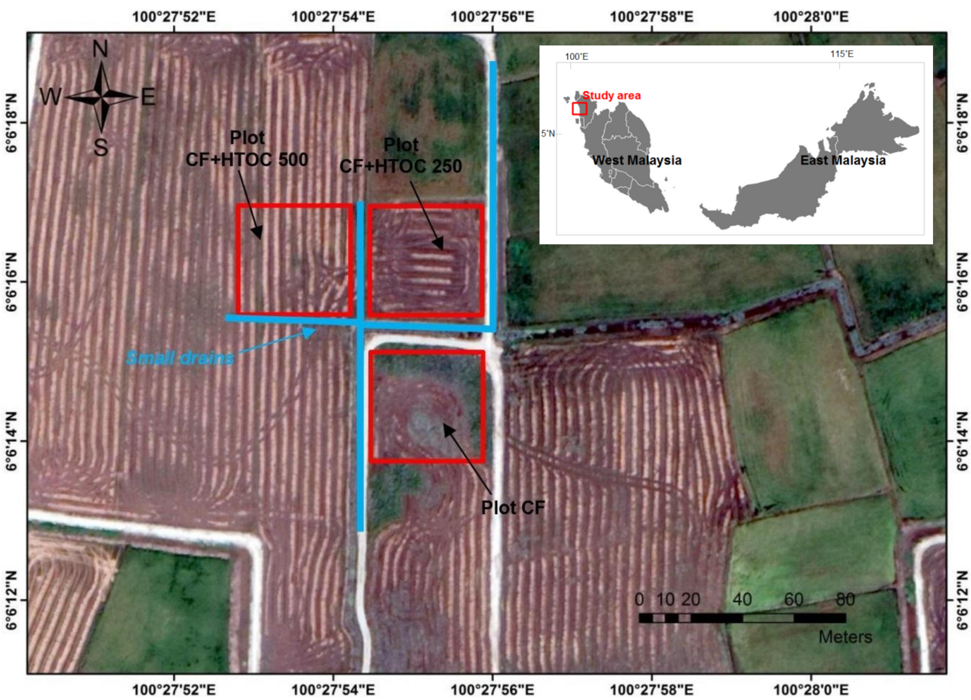

FIGURE 1. Location of the study area

with an average annual rainfall was around 2,363.9 mm. The monthly rainfall during this study was presented in Figure 2. The rainfall pattern in 2016 and 2017 was almost the same. The highest rainfall or wet season was observed in October 2016 (419.2 mm) and October 2017 (439.2 mm). While the lowest rainfall was found in March 2016 (0.40 mm) and February 2017 (17.2 mm) which was indicated as a dry season period. In this study, the sampling period for the first season was carried out in October 2016 (wet season) and the second season in April 2017 (dry season). The USDA classification has classified the soil in the study area into Inceptisols Order with Sulfic Endoaquepts group. The initial physicochemical properties of the soil is shown in Table 1.

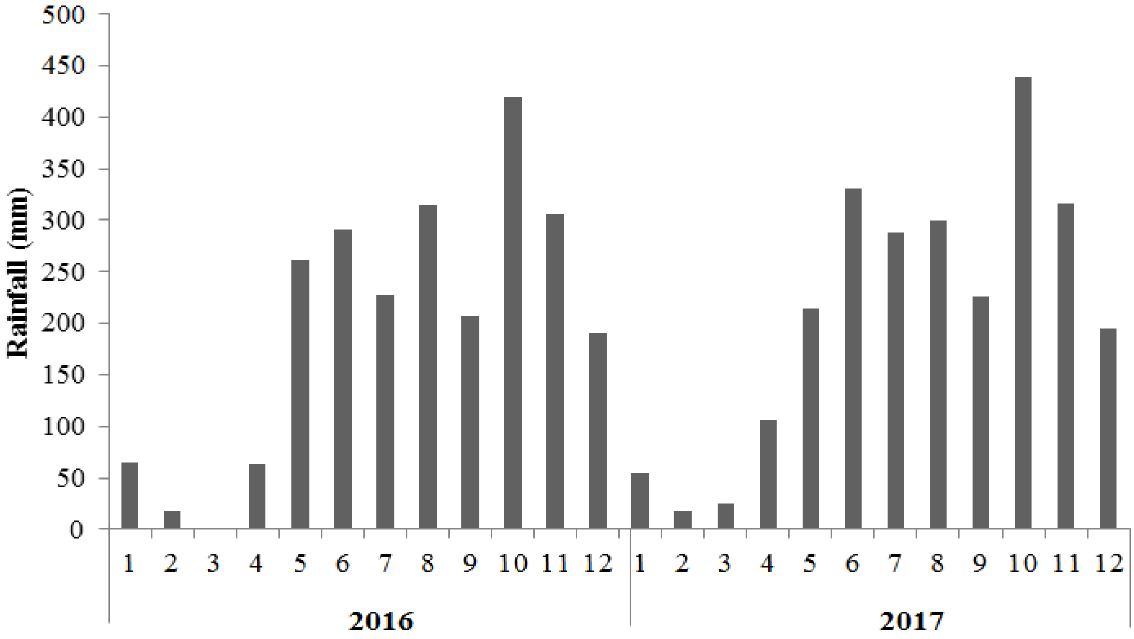

FIGURE 2. The average of monthly rainfall from 2016 to 2017 in the study area. Number 1-12 in the X axis represent the twelve months in a year

TABLE 1. The basic properties of paddy soil and organic amendment used in this study

	pH	OM (%)	C:N	Tot. Cu	Tot. Zn	Tot. Cd	Tot. Cr	Tot. Pb	Tot. As
				mg/kg					
Paddy soil	3.2	2.16	5.58	4.93	4.38	0.03	18.8	24.42	1.82
HTOC	8.1	41.69	33:1	4.85	9.39	0.07	4.32	4.30	1.28
Soil amendment standard	8[a]	>20[b]	12:1-20:1[a]	47[c]	134[c]	5[c]	200[c]	300[c]	50[c]
Soil standard	5.5-6.5[d]	4-10[e]	15:1[d]	30[f]	90[f]	0.35[f]	70[f]	35[f]	6[f]

[a]Organic fertilizer investigation level by Liu et al. (2020); [b]Recommended values of basic properties in organic fertilizer by Philippine National Standard; [c]Recommended values of basic properties in organic fertilizer by Department of Agriculture Malaysia; [d]Recommended values of soil quality in paddy soil by MARDI (2008); [e]Recommended values of soil quality in soil by Acres et al. (1975); [f]Recommended maximum concentration of heavy metal in soil by Kabata-Pendias (2011)

NPK FERTILIZER AND ORGANIC AMENDMENT

Chemical fertilizer in this trial was a compound fertilizer (N:P:K, 17.5:15.5:10/17:20:10; a standard fertilizer for paddy cultivation). The organic amendment used in this study was made from humic acid, rubber bark dust, clinoptilolite and kieserite in a specific ratio, namely HTOC. The basic properties of the HTOC were compared with the soil amendment standard (Table 1). In addition, our results showed the HTOC product had higher chemical properties than the single humic acid product (Table 2).

TABLE 2. Chemical characteristics of the single humic acid product and the HTOC

	pH	OC (%)	CEC ($cmol_c$/kg)	K^+ ($cmol_c$/kg)	N (%)	Humic acid (%)	Water content (%)
Humic acid[a]	6.3	8.17	42.80	4.5	0.24	2.60	12
HTOC (this study)	8.1	25.80	232.89	12	0.77	4.59	0.33

[a]Humic acid product investigation level by Mindari et al. (2014)

DESIGN OF EXPERIMENT

This study was carried out from May 2016 to April 2017 encompassing two planting seasons. There were three treatments in this study: (i) CF, 1,017 kg chemical fertilizer or the total amount of chemical fertilizer used for two seasons, (ii) CF+HTOC250, 1,017 kg chemical fertilizer and 250 kg HTOC and (iii) CF+HTOC500, 1,017 kg chemical fertilizer and 500 kg HTOC (Table 3). The HTOC was used as a basal application, indicating it was applied before seeds were sown. The HTOC was applied once per season, usually two weeks after the first plow was conducted. While the chemical fertilizer was applied as a topdressing. Topdressing has closely sown paddy plants and it was spread four times per season. The amount of fertilizer and the HTOC in each period were tabulated in Table 3.

TABLE 3. The amounts of fertilizer and HTOC (kg/ha) used in this study

Treatments*	Total application amount (kg)	Total organic amendment application amount (kg)	Each period total fertilizer application amount (kg)	
			2016.5.15-2016.10.21	2016.11.4-2017.4.03
CF	1,017	0	489	528
CF+HTOC250	1,267	250	489	528
CF+HTOC500	1,517	500	489	528

*CF, Chemical fertilizer; CF+HTOC250, 1,017 kg chemical fertilizer and 250 kg HTOC; CF+HTOC500, 1,017 kg chemical fertilizer and 500 kg HTOC

SOIL SAMPLING ACTIVITY

Soil samples (0-15 cm depth) were collected in each trial plot after the harvest period in each season. Total three replicates of soil samples were taken from each plot. Samples were air-dried in the drying room for seven days and crushed by the mechanical grinder. Soil samples were sieved by a 2 mm nylon sieve for soil physicochemical analysis while for heavy metals analysis, the samples were passed through a 63 μm nylon sieve.

PHYSICOCHEMICAL ANALYSIS OF THE SOIL

Soil organic matter content was analyzed by a gravimetric method based on loss on ignition (Avery & Bascomb 1982). Soil pH was analyzed using the pH meter Model DELTA 320 Mettler with a ratio of soil water (1:2.5) (Metson 1956). Humic acid content was determined by suspending soil in NaOH (0.5 mol/L) and the supernatants were obtained by centrifugation. Then, the output of extraction was acidified up to pH 2.0 using 6 N HCl. The remainder was rinsed with HCl (0.5 mol/L) and the supernatants were merged for collecting humic acid content. Available P was extracted using a molybdate-blue colorimetric method and read by Spectrophotometer Ultraviolet Model Vis UV 1201 (Murphy & Riley 1962). Available K was extracted using acetate-acetic acid ammonium 0.5 M and read by Inductively Coupled Plasma Mass Spectrometry (ICPMS). Total C and N were analyzed by a dry combustion technique using instrument LECO CR-412 Carbon Analyzer (Fujine 2014). Total heavy metals such as Cd, As, Zn, Cu, Pb and Cr were extracted using nitric acid and perchloric acid and read by Inductively Coupled Plasma Mass Spectrometry (ICPMS) (USEPA 1996).

STATISTICAL ANALYSIS

Means comparisons were statistically analyzed by Tukey's HSD test while the association between heavy metals and soil physicochemical parameters analyzed by Pearson correlation test using IBM SPSS Statistic Version 21.

RESULTS AND DISCUSSION

SOIL pH AND ORGANIC MATTER CONTENT

The application of chemical fertilizer along with HTOC showed significantly higher soil pH than the CF treatment in all seasons (Figure 3(a)). In the study area, the acidic properties of soil originated from the pyrite mineral in the soil. This notion has been found in a previous study (Rendana et al. 2019). As a result of the HTOC application, the soil pH was slowly but surely raised, culminating in the 500 kg of HTOC at the second season (Figure 3). The increase in pH was consistent with the increasing amount of HTOC applied in the soil. The low pH value at the first season might be due to the presence of jarosite mottles in the soil. The low pH was consistent with the low concentration of available P due to the presence of sulfuric layer in the soil. Furthermore, the low pH could be caused by the long-term use of N fertilizer in the study area. A study by Hao et al. (2020) found the application of N fertilizer resulted in soil acidification and they explained that base cation uptake by the plant was a primary factor in soil acidification occurred. The application of straw return could be used to alleviate soil acidification because of N fertilizer (Li et al. 2019b).

Soil organic matter (SOM) content in the treated soil (CF+HTOC) was found higher than the CF treatment in all seasons (Figure 3(b)). The highest content of SOM was obtained after applying 250 kg HTOC in the second season (8.08 %). The CF treatment showed a low level of SOM content around 2-4%, indicating this area did not apply organic materials such as compost and rice straw into the field. As a result of the HTOC application, the SOM has increased to above 7% and it was classified as an optimum level for rice cultivation. The HTOC affected humus fractions by altering the ratio C_{HA}/C_{FA} in favor of the humic acids (Nascimento et al. 2017). The SOM was a prominent factor to improve soil physical and biological properties (He & Wu 2020). Based on our study, we assumed that applying 250 kg HTOC could give an optimum result in increasing SOM, when the amount of HTOC was increased to 500 kg, the SOM content would decrease. According to Shahbaz et al. (2017), less residue protection and the increased mineralization could deteriorate the stabilization of organic matter in the soil.

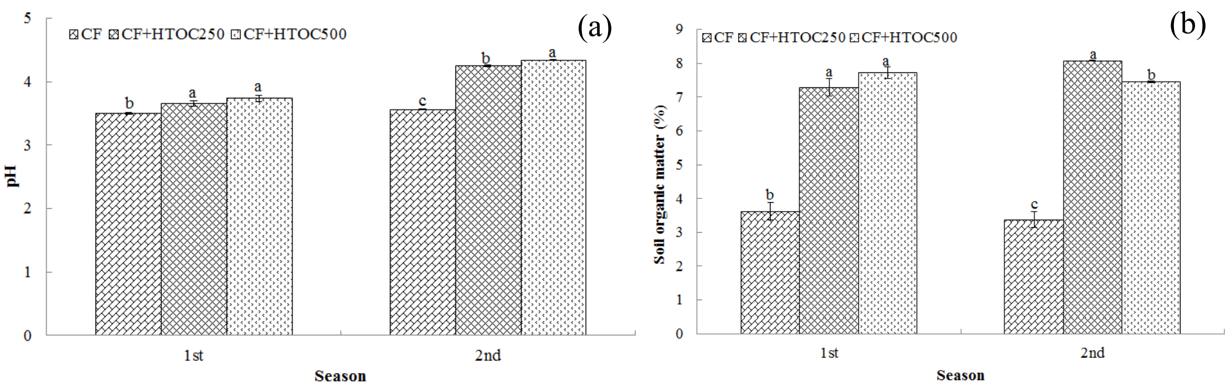

FIGURE 3. Soil pH (a) and soil organic matter content (b) in all the treatments across two seasons. Small letters represented significant differences ($p < 0.05$) among the treatments of each season

MACRONUTRIENT CONTENT

The treated soil (CF+HTOC) showed significantly higher total N than the CF treatment (Figure 4(a)). The highest total N was obtained by applying HTOC about 250 kg in the second season (0.36%). This value has been at an optimum level for rice cultivation based on classification by MARDI (2008). In contrast, the CF treatment showed the total N was very low (< 0.2%) which would affect rice growth since nitrogen was a key element for crop production. In this study, we also found there was a significant increase of total N from the first season to the second season, indicating the higher amount of HTOC applied would increase total N in the soil.

The available P in the treated soil (CF+HTOC) was significantly higher than the CF treatment (Figure 4(b)). The available P increased from the first season to the second season with optimum P content after applying 250 kg HTOC (0.65 mg/kg). Based on the classification of Acres et al. (1975), available P in all the treatments were categorized as low level (< 10 mg/kg). We assumed that two consecutive years of the HTOC application was not enough time to increase P towards optimum level because it took more time to improve soil pH which originated from strong acid soil. Several studies have found low P content related to acid sulfate soils (Phuong et al. 2020; Yli-Halla et al. 2017).

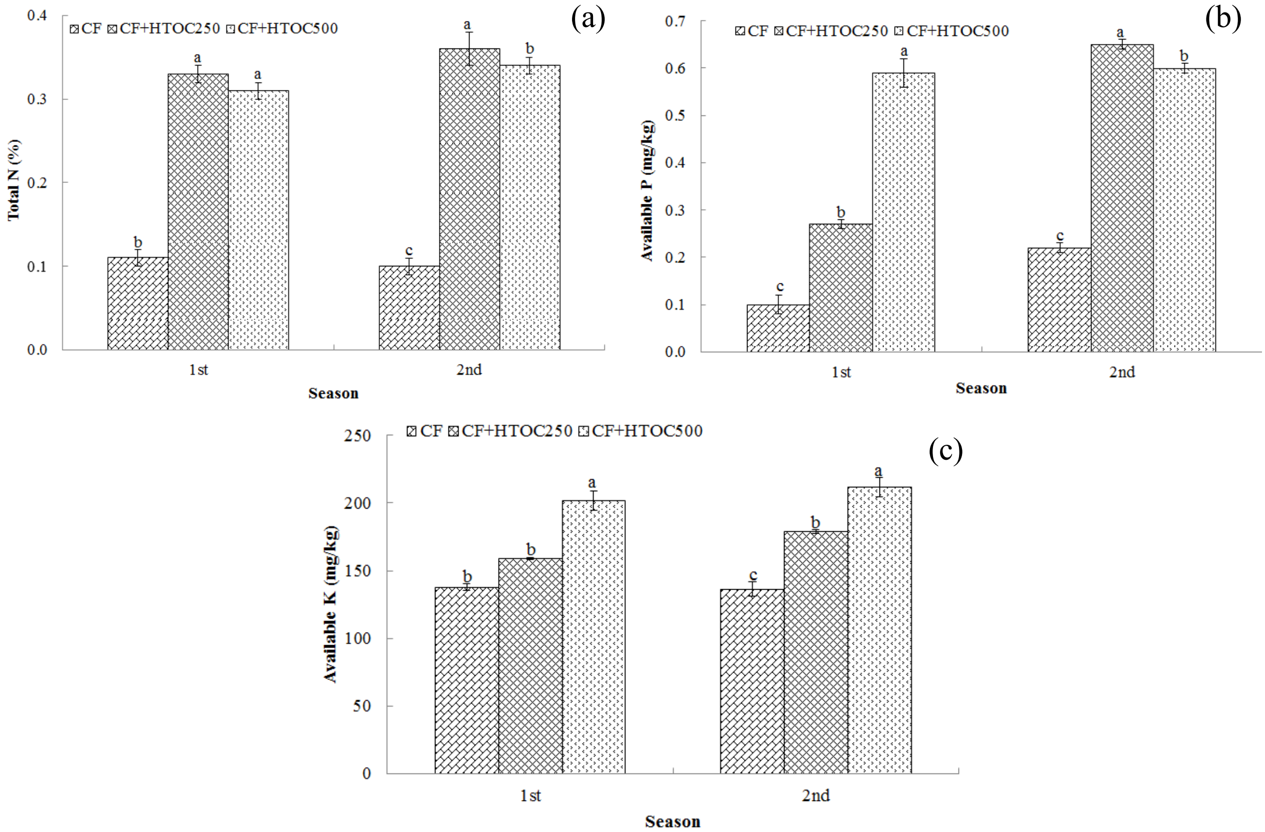

FIGURE 4. Total N (a), available P (b) and available K (c) in all treatments across two seasons. Small letters represented significant differences ($p < 0.05$) among the treatments of each season

The available K in the treated soil (CF+HTOC) significantly higher than the CF treatment (Figure 4(c)). The available K sharply increased from the first season to the second season with optimum K content after applying 500 kg HTOC (211.69 mg/kg). Overall, the available K in all the treatments has indicated a good level for rice cultivation. This result was consistent with a study by Li et al. (2019a) who found soil treated with organic amendment (mixed vermicompost and compost) showed highly increases of total N and P. The HTOC as a source of nutrient because it contained kieserite and zeolite minerals that affected considerably nutrient fractions by adding nutrient ions into the soil matrix. Mi et al. (2019) found rice grain yield for the combination of NPK fertilizer with cattle manure and rice straw were 11.4 and 9.3% higher, respectively, than the NPK alone treatment. Several studies also suggested continuous application of organic amendment could enhance soil nutrient availability through biological and enzyme activities (Chen et al. 2018; Luo et al. 2019).

HEAVY METAL CONTENT IN SOIL

The result of the study showed that the continuous application of chemical fertilizer without the addition of organic amendment might result in the accumulation of heavy metals in the soil. This was because the heavy metals were in stable forms and they tended to leach from the soil in a minor portion (Zhang et al. 2017). The application of HTOC had no significant effects on As concentration in the soil across two seasons (Figure 5(a)). Initially, the Cd concentration was found lower than the CF treatment after applying 250 kg HTOC (0.03 mg/kg), but they increased significantly after the dosage of HTOC was increased to 500 kg HTOC (0.07 mg/kg) at the first season. The optimum reduction of Cd was found at the second season where the treated soil (CF+HTOC) had a lower Cd concentration than the CF treatment (Figure 5(b)). The reduction of Cd level could occur when organic matter from HTOC made strong complexes with heavy metals. Moreover, there was a decrease in the content of Cd with an increase in pH. It was proved by a negative correlation between Cd content and pH of the soil with $r = -0.713$, $p < 0.01$ (Table 4). This result was consistent with the findings of previous studies which found the continuous application of chemical fertilizer could significantly raise the contents of Cd, Cu and Zn in soil (Huang et al. 2018; Xiaobing et al. 2020).

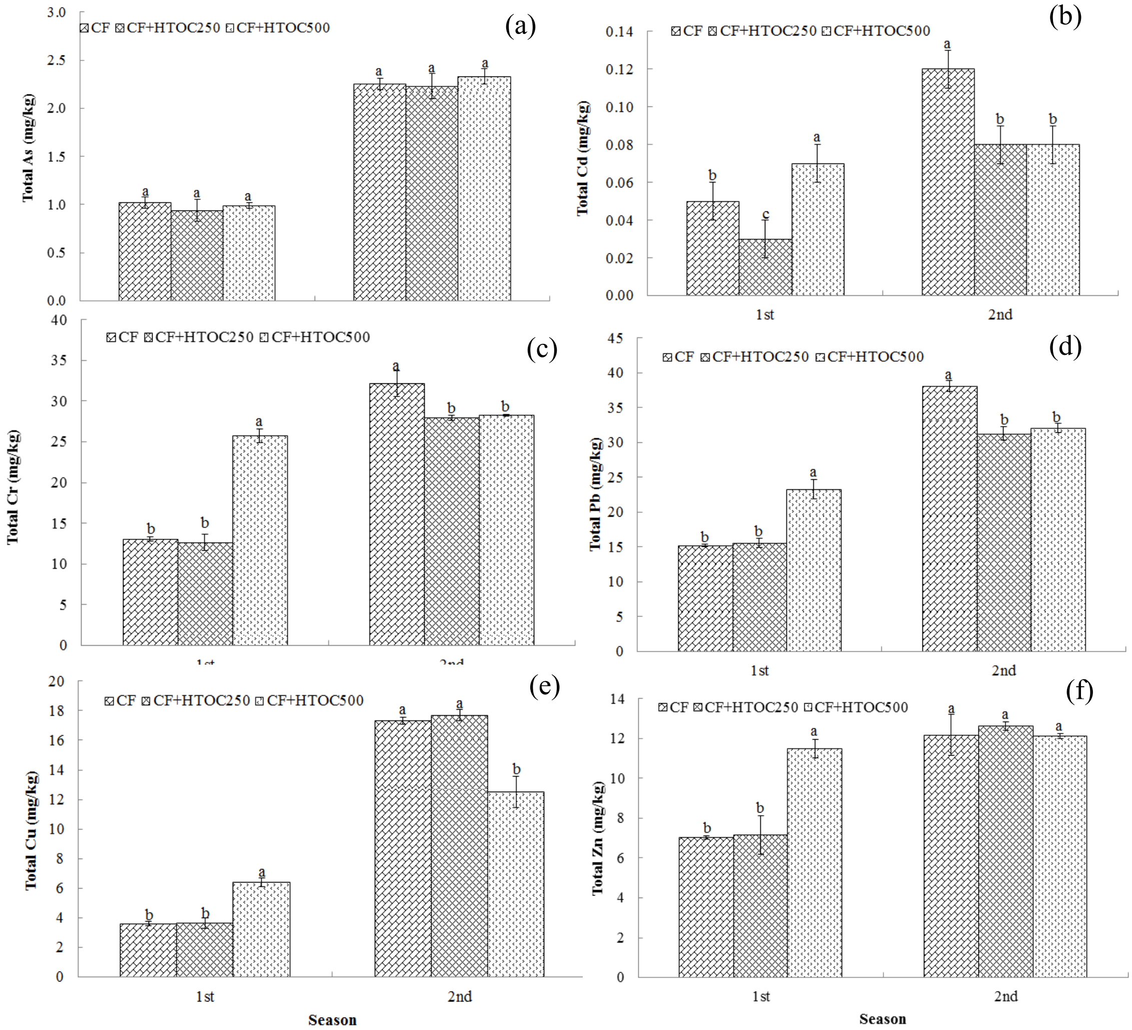

FIGURE 5. The concentrations of As (a), Cd (b), Cr (c), Pb (d), Cu (e) and Zn (f) in all treatments across two seasons. Small letters represented significant differences ($p < 0.05$) among the treatments of each season

Furthermore, the Cr and Pb concentrations were found higher than the CF treatment after applying 500 kg HTOC (25.75 and 23.27 mg/kg, respectively) at the first season. However, after continuous application, the Cr and Pb concentrations in the treated soil (CF+HTOC) were found lower than the CF treatment at the second season (Figure 5(c)-5(d)). The reduction of Cd level in the treated soil because organic matter from HTOC dominated metal (e.g Cr) binding in the soil. It was supported by a strong negative correlation between organic matter and Cr concentration with $r = -0.521$, $p < 0.01$ (Table 4). In natural conditions, high concentrations of Cr and Pb in the soil might be caused by pyrite content. The pyrite content in the soil was usually enriched with high heavy metal level like Co, Cr and Pb. The addition of HTOC has also increased the cation exchange capacity (CEC) of the soil so that the high CEC could bind heavy metal ions and had a role as a cation exchanger. The correlation analysis obtained a negative correlation between Pb ($r = -0.784$, $p < 0.01$) and Cr ($r = -0.973$, $p < 0.01$) contents with the CEC of the soil (Table 4).

Similarly, for Cu and Zn, both metals showed higher values than the CF treatment after applying 500 kg HTOC (6.39 and 11.49 mg/kg, respectively) at the first season. In contrast, in the second season, the significant reduction of Cu was observed where only the treated soil with the addition of 500 kg HTOC obtained lower Cu than the CF treatment (Figure 5(e)), while there was no significant difference found for Zn content in all the treatments (Figure 5(f)). The HTOC product tended to contain less Cu and Pb (Table 1), and it would not add Cu and Pb contents in the soil, but it could lead to Zn accumulation in the soil since the amendment had a little more Zn element. The Zinc was a micronutrient, thus it was required in small portions to sustain rice growth.

Although this amendment contributed to heavy metal accumulation in the soil, it was still lower than a critical level of the soil. The concentrations of heavy metal presented in the treated soil were still lower than the critical value by Kabata-Pendias (2011) that shown in Table 1. If we compared it with other organic compounds such as heavy metals carried by animal manures, the HTOC was still at a lower level (Yang et al. 2017). In addition, humus and organic materials like HTOC had an important role in the chelation process, diminishing and mobilizing heavy metals to plant (Dinu 2017). Based on the result of this study, we suggested using 250 kg HTOC at the first season then it was gradually increased to 500 kg HTOC at the second season to avoid heavy metal accumulation in the soil at the first season.

TABLE 4. The correlation analysis between chemical characteristics and heavy metals

	As	Cd	Pb	Cr	Zn	Cu
As	1					
Cd	0.175	1				
Pb	0.007	0.599*	1			
Cr	0.178	0.191	0.788**	1		
Zn	-0.327	-0.026	-0.269	-0.057	1	
Cu	-0.192	0.244	-0.481	-0.824**	0.233	1
pH	0.188	-0.713**	-0.746**	-0.330	0.082	-0.139
CEC	-0.163	-0.115	-0.784**	-0.973**	0.211	0.872**
SOM	-0.681**	-0.050	-0.317	-0.521**	0.321	-0.180

*Correlation is significant at 0.05 level, **Correlation is significant at 0.01 level

Conclusion

This study concluded the application of HTOC could increase soil pH, soil organic matter, total N, available P, and available K in the soil. Although the addition of HTOC could slightly contribute to higher heavy metals such as Cd, Cr, Cu, Pb, and Zn in the soil they were still below a critical limit of the soil. The heavy metal concentration in the treated soils with HTOC were high at the first season, but based on this study, it had lower levels than the chemical fertilizer treatment at the second season. As a whole, the continuous application of HTOC product could help to enhance the quality of paddy soil for rice production. For future works, this study suggests using this amendment with other agricultural activities such as oil palm, rubber, kenaf and other crops.

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